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Abstract This report describes a closed-chamber method for laboratory measurements of the rate at which radon-222 degasses (exhales) from small building material samples. The chamber is 55 L in volume and the main sample geometry is a slab of dimensions 5x30x30 cm^3. Numerical modelling is used to assess (and partly remove) the bias of the method relative to an ideal measurement of the free exhalation rate. Experimental results obtained with the method are found to be in agreement with the results of an open-chamber method (which is subject to different sources of error).

Results of radon-222 exhalation rate measurements for 10 samples of Danish building materials are reported. Samples include ordinary concrete, lightweight aggregate concrete, autoclaved aerated concrete, bricks, and gypsum board. The maximum mass-specific exhalation rate is about 20 $\mathrm{mBq\,h^{-1}\,kg^{-1}}$. Under consideration of the specific applications of the investigated building materials, the contribution to the indoor radon-222 concentration in a single-family reference house is calculated. Numerical modelling is used to help extrapolate the laboratory measurements on small samples to full scale walls. Application of typical materials will increase the indoor concentration by less than 10 $\mathrm{Bq\,m^{-3}}$.

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1 Introduction

The main objectives of this report are:

- to describe a closed-chamber method used at Risø for laboratory measurements of the radon-222 [1] exhalation rate of building materials,
- to investigate the various sources of errors characteristic for this method,
- to report results for 10 Danish building material samples, and
- to extrapolate the results to a typical Danish single-family house.

The report includes a brief review of other methods for exhalation rate measurements, as well as results of previous exhalation-rate measurements conducted in Denmark. The company H+H Industri A/S, Ølsted, Denmark, has supported the present work financially. Furthermore, all building-material samples have been selected and supplied by that company, and most (but not all) of the materials were produced there.

H+H Industri A/S

1.1 Background

Danish homes without direct ground contact (for example, apartments in multi-story buildings) typically have a low indoor radon level of about 20 $\mathrm{Bq\,m^{-3}}$ [SIS87^b]. In comparison, the average indoor radon level in Danish single-familiy houses is normally about 70 $\mathrm{Bq\,m^{-3}}$, and the level can be very different from house to house. For example, single-family houses with annual averages from 10 to 1000 $\mathrm{Bq\,m^{-3}}$ have been found [An97^b]. The reason for the pronounced difference between houses with and without direct ground contact is that soil gas has a high concentration of radon (typically about 10 000 to 100 000 $\mathrm{Bq\,m^{-3}}$). Even a minute entry rate of soil gas therefore can have a large impact on the indoor radon concentration. Such gas entry is possible because most houses (apparently) do not have a gas-tight floor construction and because houses are normally at a slight underpressure (as it is normally warmer indoors than outdoors). Soil is therefore the main source of indoor radon in most Danish single-family houses [SIS87^b, An97^a].

Soil-gas entry

In line with the above, studies carried out by Jonassen, Ulbak and co-workers in the 1970'ies and 1980'ies (see Section 8.2), showed that radon exhalation rates of ordinary Danish building materials are low. This is confirmed by the present investigation. Special building materials with large radon exhalation rates do however exist (at least in other countries): alum-shale concrete, granite, Italian volcanic tuff, and by-product gypsum[2] [UN93]. For example, Sweden has about 300 000 houses with alum-shale building materials sometimes referred to as "blue concrete" a lightweight aerated concrete used in blocks [SSI93]. This building material can raise the indoor radon level above 1000 $\mathrm{Bq\,m^{-3}}$. Alum-shale concrete is no longer produced, and its application in Denmark has been limited [Ul80]. Finally, it should be observed that soil can have a large emanation rate (e.g. above 10 $\mathrm{atoms\,s^{-1}\,kg^{-1}}$ which is about five times the mass-specific exhalation

Building materials

[1] The most abundant radon isotope is radon-222. It ordinates from radium-226 and is part of the Uranium Series (U-238). The half life of radon-222 is 3.83 days. Because of the importance of this particular isotope, it is often referred to as "radon". As this report concerns only radon-222 (and not radon-220 or any other isotope of radon) we will adopt this practice here.

[2] Phosphogypsum is used in some countries as a substitute for natural gypsum in the manufacture of cement, wallboards and plaster [UN93]. Phoshogypsum is a by-product from the fertilizer industry. The elevated levels of radium-226 in phospogypsum comes from the phosphate rock that tends to have elevated concentration of Uranium-238 decay products.

rate for ordinary concrete). Therefore, so-called "ecological" houses build in Denmark from clay excavated directly on the building site may have building materials with above-average radon exhalation rates.

The Danish radon budget

Budget 1 In an investigation of radon in 117 newer Danish single-family houses [An97[a]], it was found that radon entered at a (geometric) mean rate of 9.6 $\mathrm{kBq\,h^{-1}}$ and that 80 % of the houses had radon entry rates above 6 $\mathrm{kBq\,h^{-1}}$. In the typical Danish single-family house built of clay bricks and/or aerated concrete, the radon entry rate resulting from building materials has been estimated to be 1–3 $\mathrm{kBq\,m^{-3}}$ [Jo80, Ul84]. Entry from the outdoors amounts to about 2 $\mathrm{kBq\,h^{-1}}$ or less. Hence, this calculation suggests that building materials and outdoor air cannot account for observed indoor radon levels in 80 % of the investigated houses.

Budget 2 Another "radon budget" comes from the results of the 1985–86 national survey [SIS87[b]]: The average radon level in multi-family houses was found to be 19 $\mathrm{Bq\,m^{-3}}$. Since the outdoor radon level is about 8 $\mathrm{Bq\,m^{-3}}$ [Ma86], and since entry from the soil is probably marginal for most of the multi-family houses included in the survey, it seems reasonable to attribute $19-8 = 11$ $\mathrm{Bq\,m^{-3}}$ to radon from building materials.

1.2 Organization of the report

Section 2 gives a presentation of the theoretical framework for radon exhalation. Quantities such as the mass-specific exhalation rate are defined, and it also outlines the characteristics of various methods for exhalation rate measurements. Section 3, includes descriptions of samples as well as the experimental apparatus. In Section 4, the experimental procedure is described, and it is shown how the final exhalation rate results are found from raw measurements. The results of the exhalation rate measurements are given in Section 5. Section 6 contains the results of numerical model calculations. Based on solution of the 3-dimensional time-dependent diffusion equation, a number of issues related to the measurement procedure are investigated. A reference house is then defined in Section 7, and under consideration of the various applications of the building materials, the concentration in such a house is estimated. The final two sections of the report contain discussion and conclusions. An appendix contains measurement sheets for all exhalation-rate measurements.

2 Theoretical framework

2.1 Radiometric quantities

Exhalation rates, J

Consider a certain sample of building material placed in some well-defined environment of given temperature, humidity, pressure, stress, radon concentration etc. Under the given conditions, we define the sample specific exhalation rate J to be the net amount of radon that escapes the sample per time unit.

Main units In this report 'the amount of radon that escapes per time unit' is expressed as the number of radon atoms that escapes per second (atoms $\mathrm{s^{-1}}$), or as the amount of radon activity measured in Bq that escapes per hour ($\mathrm{Bq\,h^{-1}}$). From the basic

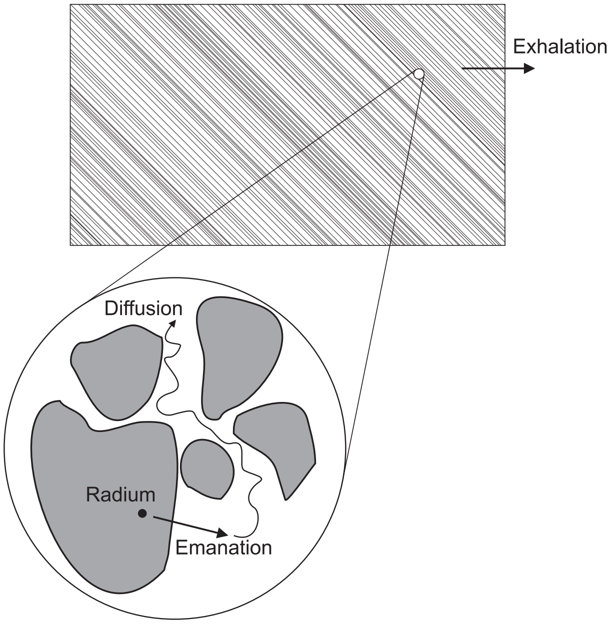

Figure 1. Illustration of the main processes involved: 1) radium inside a grain decays to radon, 2) some of the radon atoms reach the pores of the material (this is called emanation), 3) radon diffuses through the pore system, and 4) part of the pore radon degasses from the surface of the material (this is called exhalation).

law of radioactive decay, we have that:

$$[J \text{ in units of Bq h}^{-1}] = \lambda \, \frac{3600 \text{ s}}{1 \text{ h}} \, [J \text{ in units of atoms s}^{-1}] \tag{1}$$

where λ=2.09838 $\cdot\, 10^{-6}$ s^{-1} is the decay constant of radon. Hence, the statement that a sample has an exhalation rate of 1.0 atoms s^{-1} is equivalent to the statement that the sample has an exhalation rate of 7.6 $\cdot\, 10^{-3}$ Bq h^{-1}.

If A is the total geometric surface area of the sample and M is the mass of the sample, we then calculate the area specific (J_{A}) and the mass specific (J_{M}) radon exhalation rates as:

$$J_{\mathrm{A}} = \frac{J}{A} \tag{2}$$

$$J_{\mathrm{M}} = \frac{J}{M} \tag{3}$$

In this report J_{A} is expressed in units of atoms s^{-1} m^{-2} or Bq h^{-1} m^{-2}. Likewise, J_{M} is expressed in units of atoms s^{-1} kg^{-1} or Bq h^{-1} kg^{-1}.

As already indicated, the exhalation rate of a given sample depends on the environment in which the sample is placed. The situation when the environment has zero radon concentration is of special interest. We refer to this situation as "free", and add the letter 'f' as a subscript to exhalation rate quantities obtained under this condition ($J_{\mathrm{M,f}}$ and $J_{\mathrm{A,f}}$). Likewise, so-called bound exhalation rates (defined page 5) are given the letter 'b' as subscript (e.g. $J_{\mathrm{M,b}}$ and $J_{\mathrm{A,b}}$).

Free exhalation

Bound exhalation

Radium concentration, A_{Ra}

Radium-226 is transformed to radon by radioactive decay. Therefore, radon is produced in all materials containing radium-226. The concentration of radium A_{Ra} in units of Bq radium-226 per kg dry mass directly gives the production rate of radon. For example, if a sample contains 23 Bq radium-226 then it means that radon is produced at a rate of 23 atoms per second. The radium content of the building materials depend solely on the selected raw materials.

Radon emanation rate, E

In porous materials, radium is situated in solid grains. Not all radon produced in the grains actually escape to the pores in between grains. We define the radon emanation rate E to be the number of radon atoms per second per kg dry material (atoms s^{-1} kg^{-1}) that escape the solid parts of the material and are available for transport at a scale larger than the characteristic pore diameter of the material. Essentially, the emanation rate is the rate at which radon is supplied to the pores of the material.

Fraction of radon emanation, f

The fraction of emanation, f, is here defined as the ratio between the rate of emanation, E, and the rate of radon production inside the sample (i.e. the radium-226 concentration, A_{Ra}):

$$f = \frac{E}{A_{\mathrm{Ra}}} \tag{4}$$

The fraction of emanation depends on the distribution of radium-226 in the grains, the grain size distribution and the presence of moisture in between grains. Theoretically, f may take values from 0 to 100 %. For example, a large fraction of emanation can be expected if radium exists as a surface coating on the grains, if the grains have a large inner porosity, or if the grains are very small. Presence of water in between grains can also moderate the emanation process [Ta80]. For soils, the typical maximum value of the emanation fraction is about 20 %. For a given material (with fixed grain size distribution etc.) the emanation fraction is essentially only a function of the moisture in the sample. It is assumed, that f is independent, for example, of the radon concentration in the pores.

Influence of moisture

Fraction of exhalation-to-emanation, g

Radon can move through the sample by diffusion and advection. Because of the finite half-life of radon, only a fraction of the pore-space radon escapes the sample before decay. We introduce the fraction of emanation-to-exhalation, g, as:

$$g = \frac{J_{\mathrm{M}}}{E} \tag{5}$$

where J_{M} is the mass-specific exhalation rate and E is the emanation rate. g takes values from 0 to 100 %. If the sample is very large, only a small fraction of the radon generated inside the sample will reach the surface. In that case, g will be close to zero. If the sample is very small, all radon generated in the sample will probably reach the environment and g is unity.

Pressure differences across the sample can induce flows of air which in turn transport radon advectively. The main source of bulk air movement through intact samples (i.e. samples without macroscopic cracks) are probably changes in the absolute atmospheric pressure.

Advective transport

Fraction of radon exhalation, h

We define the fraction of exhalation, h, as the ratio between the rate of radon exhalation from the sample and the rate of radon production inside the sample (i.e. the radium concentration):

$$h = \frac{J_{\mathrm{M}}}{A_{\mathrm{Ra}}} \tag{6}$$

h takes values from 0 to 100 %. As already described, the process of exhalation can be split into two parts as described by f and g, and we have:

$$h = \frac{J_{\mathrm{M}}}{E} \cdot \frac{E}{A_{\mathrm{Ra}}} = f \cdot g \tag{7}$$

Diffusivity, D

Exhalation of radon from building materials such as concrete mainly results from molecular diffusion [St88, Rog94, Re95]. The bulk diffusivity, D, of building materials is therefore an important parameter. D is normally believed to be in the range from 10^{-10} m^2 s^{-1} to 10^{-6} m^2 s^{-1}, and this is also the range considered in the model calculations in Section 6.

The diffusive flux is proportional to the gradient of the radon concentration field. To clarify what this means, we consider the following example (a more formal definition of bulk diffsuivity can be found elsewhere–see e.g. [An92, An99]): Imagine a A=120 m^2 house positioned on soil. The house has an intact slab of L =0.1 m in thickness. The slab has a bulk diffusivity D=10^{-6} m^2 s^{-1}. The radon concentration below the slab is set to be c_A=50 000 Bq m^{-3}. The indoor environment has a near-zero radon concentration c_B. Ignoring radioactive decay, the diffusive entry J of radon to the house is:

Diffusion through a concrete slab

$$J = AD\frac{c_A - c_B}{L} \tag{8}$$

$$= 120\ \mathrm{m}^2 \cdot 10^{-6}\ \mathrm{m}^2\ \mathrm{s}^{-1} \cdot \frac{50\ 000\ \mathrm{Bq\,m}^{-3}}{0.1\ \mathrm{m}} \tag{9}$$

$$= 21\ 600\ \mathrm{Bq\,h}^{-1} \tag{10}$$

If the house has an air-exchange rate of λ_{v}=0.5 h^{-1} and a volume of V=300 m^3, then the diffusive entry rate can increase the indoor radon concentration by as much as 173 Bq m^{-3} (see mass-balance model described page 25). If the bulk diffusivity of the slab is 10^{-10} m^2 s^{-1}, the diffusive entry through the slab can not even increase the indoor radon concentration by 1 Bq m^{-3}.

Concentration field reshaping

The diffusive exhalation rate from a sample is always at a maximum when the sample is placed in a zero-concentration environment. This is referred to as free exhalation (J_{f}). If the sample is placed in a closed chamber (with no other sources of radon), and if the chamber is initially at zero concentration, then initially (i.e. at $t = 0$) radon will exhale from the sample at a rate corresponding to the free exhalation rate, $J(0) = J_{\mathrm{f}}$. Because the chamber is closed, the radon concentration will inevitable increase as a result of this. This leads to a new (less steep) radon concentration profile in the sample–i.e. the field is reshaped [Sa84]– and the exhalation rate decreases (i.e. $J < J_{\mathrm{f}}$ for $t > 0$). If the chamber is small compared with the sample, the change in exhalation rate can be large. If no changes are made to the system, the radon concentration in the chamber will approach some equilibrium value, c_∞. At that point, the net exhalation from the sample is balanced by radioactive decay of radon in the air volume of the chamber and leakage of radon out of the chamber. The exhalation rate at this point is called 'the bound exhalation rate', and we use the subscript b to mark this condition. Hence, we use J_{b} for the bound sample-specific exhalation rate, $J_{\mathrm{A,b}}$ for the bound area-specific exhalation rate, and $J_{\mathrm{M,b}}$ for the bound mass-specific exhalation rate.

Free exhalation

Bound exhalation

From exhalation rate measurements to full-scale walls

An important application of laboratory measurements of the exhalation rate of (small) samples of building materials is to assess the contribution of those materials when applied in specific house-construction parts. This is discussed in Section 6.5.

2.2 Measurement procedures

Measurement of radon exhalation rates can be performed in a multitude of ways. The most important ones are outlined below.

Gamma measurements of the radium-226 content

Radium-226 is the source of radon. A crude (but robust) measure of 'potential radon exhalation' is to obtain the radium-226 concentration A_{Ra} ($\mathrm{Bq\,kg^{-1}}$) of the material. Such a measurement can be performed by gamma spectroscopy. From the conservative assumption, that all radon generated inside the building material gets out (i.e. assuming h=1.0 in equation 7), we have:

$$J_{\mathrm{M}} = A_{\mathrm{Ra}} \tag{11}$$

Radium index

With further assumptions, it is even possible to put an upper bound on the indoor radon level. For example, building materials complying with the Swedish radium index requirement:

$$A_{\mathrm{Ra}} < 200\ \mathrm{Bq\,kg^{-1}} \tag{12}$$

cannot raise the indoor radon level by more than about 150 $\mathrm{Bq\,m^{-3}}$ even if the floor, ceiling, and all house walls are made with that material [Cl92, p. 102]. The house is set to have an air-exchange rate of 0.5 $\mathrm{h^{-1}}$.

Gamma measurements of the above type are relatively easy to conduct, but do not give the actual rate of radon exhalation.

Laboratory measurements of radon exhalation

Laboratory measurements are conducted by placing the sample under investigation in a chamber from which the radon concentration can be measured. The main two measurement procedures can be outlined as follows:

Open-chamber method (Method A) A flow of air (typically about 1 $\mathrm{L\,min^{-1}}$) is established through the chamber. This provides the sample with a well-defined environment with respect to humidity and radon. A near-zero radon concentration is preferable for measurements of the free exhalation rate (see page 5). After a selected time of conditioning (e.g. 12 h), the sample is assumed to be in equilibrium with the chamber environment, and radon measurements are conducted. There are now two ways to proceed:

Activity collection (Method A1) A device able to trap radon is placed at the outlet of the chamber. This can for example be a cold trap of activated charcoal placed in a dewar with dry ice (temperature −78 °C). Such a trap will effectively collect all radon leaving the chamber. The trapped activity subsequently can be determined by gamma spectroscopy. Alternatively, radon may be released into a scintillation cell [Ma88]. If the activity A (Bq) is trapped over a period of time T (s), then the exhalation rate J ($\mathrm{Bq\,s^{-1}}$) from the sample is:

$$J = \frac{A}{T} \tag{13}$$

The main feature of this approach is that only the determination of A is a subject to uncertainty. The experimenter even has the opportunity to diminish the counting error relating to the A-determination by selecting a sufficiently long time of integration. This method therefore is (potentially) very accurate and highly sensitive.

Air concentration measurement (Method A2) A sample of air is taken from the chamber outlet or the radon concentration of the chamber is monitored continuously. From the measured concentration c ($\mathrm{Bq\,m^{-3}}$) and the flow rate Q ($\mathrm{m^3\,s^{-1}}$) through the chamber, the exhalation rate J ($\mathrm{Bq\,s^{-1}}$) can then be found as:

$$J = cQ \tag{14}$$

Since the radon concentration is low (typically below 5 $\mathrm{Bq\,m^{-3}}$), this method is only useful with a sensitive method for radon concentration determination. In most cases, counting error will be an important source of uncertainty. Another source of error is in the flow rate determination.

The open-chamber method A1 follows the general recommendations given by the Danish Standards Association regarding degassing measurements for building products [DS94]. Another reason to consider method A1 a good reference method is that it follows the principles in the proposed Dutch norm for exhalation rate measurements (the pre-standard is identified as NVN5699). It is used for example by the KVI [Gr97]. *Standard methods*

Closed-chamber method (Method B) First the sample is conditioned as described with the previous method. If the flow rate of air through the chamber is sufficiently large (and is without radon), the chamber will quickly approach a near-zero level. At time $t = 0$, the chamber is closed in the condition:

$$c(0) \approx 0 \tag{15}$$

As a result of exhalation from the sample, the radon concentration starts to build up inside the chamber for $t > 0$. Monitoring of the radon concentration $c(t)$ in the chamber over a certain period of time (e.g. 5–30 days) is done by grab sampling or by a continuous radon monitor. The analysis of the so called 'growth curve' $c(t)$ is conducted as follows: If there are no leaks in the chamber (i.e. if the chamber is truly closed), mass balance requires that:

$$V\frac{\mathrm{d}c}{\mathrm{d}t} = J(t) - \lambda V c \tag{16}$$

where we have assumed that the chamber is well mixed. V is the volume of the chamber. If the chamber is sufficiently large (compared with the sample) it is a good approximation (see section 6) to assume that the exhalation rate is constant:

$$J(t) \approx J \tag{17}$$

such that equation 16 has the solution:

$$c(t) = c_\infty(1 - e^{-\lambda t}) \tag{18}$$

where

$$c_\infty = \frac{J}{\lambda V} \tag{19}$$

is the radon concentration ($\mathrm{Bq\,m^{-3}}$) of the chamber as $t \to \infty$. Fitting equation 18 to the measured growth curve $c(t)$ provides an estimate of the parameter c_∞ from which the exhalation rate can be found as:

$$J = \lambda V c_\infty \tag{20}$$

This method is critically dependent on the assumption that the chamber is leak free. If this is not the case, λ in equation 20 needs to be substituted by some effective 'decay constant' λ_{eff} that incorporates radioactive decay as well as the leakage. λ_{eff} can be made part of the fitting procedure described above. The correction is only valid if the (leaky) chamber is placed in a room with a radon concentration much lower than that of the chamber.

3 Materials

This section describes the investigated samples and the experimental apparatus.

3.1 Samples

Two batches of each 10 building material samples were supplied by H+H Industry A/S. Most (but not all) samples were produced by that company. The first batch was delivered to Risø on September 29, 1997. The second batch was delivered on November 10, 1997. Both batches were produced 1–2 months prior to the dates of delivery. At H+H Industry A/S, all samples had been conditioned to be in equilibrium with air at 23 °C and 43 % relative humidity. This means that the moisture content W in all samples were less than 3 %. W is the mass of (removable) moisture divided per dry mass. At Risø, the samples were located in a basement laboratory room in building 125. The typical conditions of that room were 24 °C and 40 % relative humidity. The average radon concentration in the room was about 30 Bq m^{-3}.

The samples in batch 1 are identified as M1 to M10 (i.e. material no. 1, material no. 2 etc.). Measurements were not performed for batch 2.

For the samples in batch 1, Table 1 gives linear dimensions, masses (M), surface areas (A), volumes (V), area-to-volume ratios (A/V), and densities ($\rho_{\mathrm{m}} = M/V$).

Sample M10 is an aggregate (single grains), and the volume has been calculated for a relatively loose packing. The corresponding surface area has been calculated from an assumed area-to-volume ratio of 0.533 m^{-1}. This ratio is identical to that obtained for a slab of dimensions 30 x 30 x 5 cm^3.

3.2 Equipment

Figure 2 shows the experimental set-up.

Chamber

All measurements were performed in a cylindrical stainless-steel chamber. The volume of the chamber is 55.76 L (about 34.4 cm diameter and 60 cm depth). The lid of the chamber is sealed with an o-ring and is closed by 16 bolts. The chamber is equipped with two fans of the type used for cooling in personal computers.

Flow control

The flow system consists of a dry line and a wet line. The dry-line flow comes from a 4 m^3 nitrogen (pressurized) gas cylinder. The flow rate is regulated manually by use of the pressure reduction valve. The flow has a relative humidity of 0 %. The wet-line flow comes from another 4 m^3 nitrogen gas cylinder. This flow is controlled by a mass-flow controller (Brooks Instrument B.V., the Netherlands)

Table 1. Dimensions of the samples in batch 1. The densities given in the second column are nominal factory densities in units of kg m^{-3}. Lightweight aggregate concrete and autoclaved aerated concrete are abbreviated as LAC and AAC, respectively.

ID	Description	Dimensions	Mass kg	Area m^2	Volume L	A/V m^{-1}	Density kg m^{-3}
M1	LAC, density 600	1 slab 30.0 x 30.0 x 4.9 cm^3	2.89	0.239	4.41	54	656
M2	LAC type 1, density 1500	1 slab 29.8 x 30.0 x 5.4 cm^3	7.32	0.243	4.83	50	1516
M3	LAC type 2, density 1500	1 slab 29.8 x 29.9 x 5.0 cm^3	7.04	0.238	4.46	53	1579
M4	AAC, density 450	1 slab 30.1 x 30.0 x 5.5 cm^3	2.56	0.247	4.97	50	515
M5	AAC, density 650	1 slab 30.0 x 30.1 x 5.1 cm^3	3.12	0.242	4.61	53	677
M6	AAC, density 735	1 slab 29.9 x 30.1 x 5.1 cm^3	3.66	0.241	4.59	53	797
M7	Ordinary concrete, density 2300	1 slab 29.9 x 30.0 x 5.0 cm^3	10.08	0.239	4.49	53	2248
M8	Gypsum board	5 boards 29.8 x 29.8 x 1.3 cm^3	3.96	0.963	5.55	173	713
M9	Bricks	3 bricks 10 x 20 x 5.4 cm^3 2 bricks 10 x 14.8 x 5.4 cm^3	8.56	0.330	4.84	68	1768
M10	Lightweight expanded clay aggregate	Single grains (no packing)	1.51	(0.277)[a]	5.19	(53)[a]	291

[a] See text

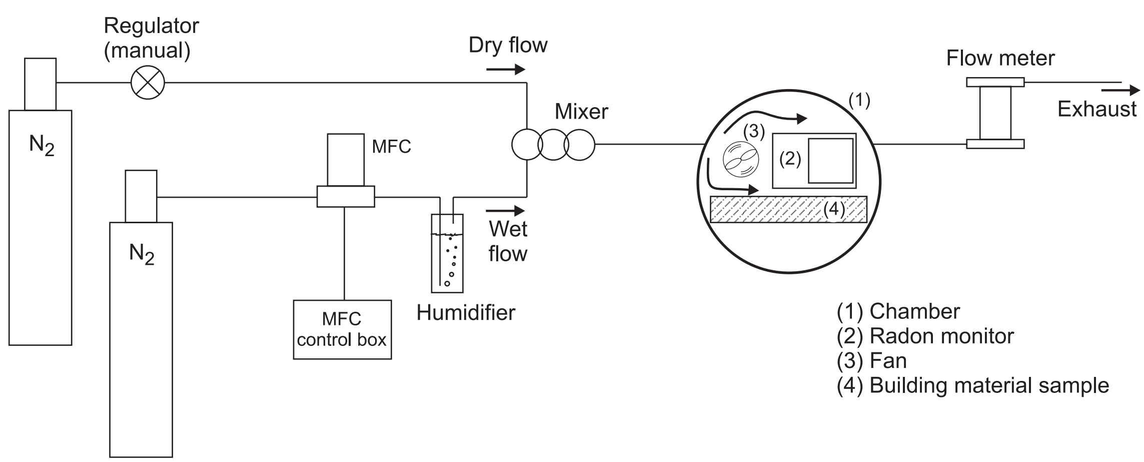

Figure 2. Experimental set-up.

set to 500 mL_n min^{-1} (nL = normal liter). The flow is led through a humidifier such that the flow thereafter can be assumed to have a relative humidity of 100 %.

The two flow lines are mixed and the flow is supplied to the chamber. The total flow rate from the chamber was measured with a Gilian bubble flow meter (20 mL min^{-1}–6 L min^{-1}; Gilian Instruments Corp. USA).

Radon instrument

Radon concentration measurements were conducted with an ionization chamber placed inside the chamber (AlphaGuard, PQ-2000 from Genitron, Germany). The monitor has sensors for relative humidity, temperature and absolute pressure. It is assessed that about 1.55 L of the monitor is rigid. This value is used for the calculation of the free air volume in chamber.

Numerical model

A numerical finite-difference model called `RnMod3d` developed at Risø was used in certain parts of the error analysis. `RnMod3d` is a 3D time-dependent model of gas and radon transport through porous media. The principles behind the model are outlined in [An92]. The model has been compared with other models [An99], and it has also successfully been tested against the analytical steady-state solutions given by Berkvens et al. [Be88].

4 Experimental procedures and data analysis

An experimental procedure corresponding to the closed-chamber method (Method B) described page 7 is adopted as primary method in this work. A (less accurate) version of the open-chamber method (Method A2) is used to check for gross errors. To distinguish between results obtained with the two methods, all open-chamber results are marked with an OC (Open Chamber) as in $J_{\mathrm{f,OC}}$.

4.1 Experimental procedures

1. The sample was weighted and positioned in the chamber.
2. From the computer, the continuous radon monitor was set to store results in a new data file. The cycle time of the monitor was set to 1 hour (preferable) or 10 min.
3. The continuous radon monitor was placed in the chamber.
4. The lid was put on the chamber.
5. The flow was started, and the time was noted in the log book as the start of conditioning. The mass-flow controller was set to 0.5 L min^{-1}. The reduction valve of the dry-flow line was adjusted such that the total flow Q leaving the chamber was about 1 L min^{-1}. The flow was maintained at this level for 12–24 hours. The total flow rate was manually measured at selected times with the bubble flow meter.
6. The tubing was removed from the chamber, and the chamber was closed. The time was noted in the log book as the time the conditioning stopped (and the build-up started).

7. After 2–14 days of build-up, the chamber was opened. The time was noted in the log book as the time the build-up stopped.

8. The sample was removed from the chamber and was (re)weighted.

9. The data from the experiment was stored in the database.

4.2 Data and error analysis

Radon monitor bias

The raw radon concentrations reported by the monitor are corrected for instrument bias by subtraction of 14.12 ± 0.72 Bq m^{-3}. Hence, a (raw) instrument reading of 15.12 ± 0.5 Bq m^{-3} is corrected to 1.0 ± 0.9 Bq m^{-3}, where all indicated (statistical) uncertainties are expressed as one standard deviation, and where the uncertainty of the corrected result is found by quadrature summation. This correction was deduced from one single blank experiment conducted from August 11 to August 13, 1998. The radon monitor was left in the chamber (without any sample), and the chamber was flushed with nitrogen let through an activated charcoal cold trap [Ma88]. Such a trap is known to effectively remove any radon in the nitrogen. To allow for desorption of radon from chamber walls, the chamber was flushed on two occasions.

The radon monitor is calibrated against three local standards all traceable to NIST (see [An97^b]). The uncertainty of the bias of the results (expressed as one relative standard deviation) is judged to be about 5 %.

Closed-chamber method (Method B)

Equation 18 was extended with a constant term c_0 and an effective decay constant λ_{eff}:

$$\hat{c}(t) = \begin{cases} c_0 \text{ for } t < 0 \text{ (conditioning)} \\ c_0 + c_\infty(1 - e^{-\lambda_{\text{eff}} t}) \text{ for } t \geq 0 \text{ (build-up)} \end{cases} \tag{21}$$

c_0 reflects the (potential) off-set of the radon monitor and the fact that the radon concentration of the air in the chamber during conditioning was only near zero but not exactly zero. λ_{eff} was set to fixed values (see later) and was not made part of the fitting procedure. A value of λ_{eff} greater than the radioactive decay constant of radon (λ=$2.09838 \cdot 10^{-6}$ s^{-1}) means that the chamber is leaky.

Non-linear fitting

Equation 21 was fitted to the measured radon concentration in the chamber during conditioning and build up. Non-linear curve fitting was conducted with the Marquard method as described in Bevington and Robinson [Be92, p. 161]. Essentially, the fitting procedure determines the values of c_0 and c_∞ such the sum-of-squares:

$$\chi^2 = \sum_{i=1}^{N} \left(\frac{c_i - \hat{c}(t_i)}{\sigma_i} \right)^2 \tag{22}$$

becomes minimal. c_i is the radon concentration measured at regular time intervals t_i (every hour or every 10 minutes), N is the number of measurement points and σ_i is the uncertainty associated with each c_i as estimated by the radon monitor. The reduced-χ^2 (χ^2_ν) is calculated as:

$$\chi^2_\nu = \frac{\chi^2}{\nu} \tag{23}$$

where $\nu = N - 2$. A value of χ^2_ν very different from unity indicates that either the fitting function or the error estimates σ_i are inappropriate.

Corrections

The free exhalation rate was calculated using a modified version of equation 20:

$$J_f = \alpha \lambda_{eff} V c_\infty \tag{24}$$

where V is the air volume of the chamber[3] and c_∞ is the fitted equilibrium radon concentration. The factor α converts the measured bound exhalation rate to the free exhalation rate J_f. Based on the results of model calculations presented page 20, α was in all cases set to $1/0.987 = 1.013$. As discussed page 28, λ_{eff} was set to $1.037 \cdot \lambda$ for measurement 103 to 120, and $1.0 \cdot \lambda$ for measurement 121, where λ is the (true) decay constant of radon ($2.09838 \cdot 10^{-6}\ s^{-1}$). The statistical variability $u\{J_f\}$ of any J_f-determination is found by quadrature summation of the following contributions:

Uncertainty analysis

- The statistical error associated with the fitted parameter c_∞ (the inverse of the diagonal element in the error matrix).
- Although measurements numbers 103 to 120 are corrected for leakage from the chamber, this leakage was probably different from experiment to experiment. The variability from this source on the final result (J_f) is judged to be about 2 % [4].
- The correction α from bound to free exhalation is set to be about 0.4 % (half the maximum range of the results shown in Figure 9). Hence: $\alpha = 1.013 \pm 0.004$, where the uncertainty is expressed as one standard deviation.
- Other (random) sources of errors (such as sink effects, interference of radon-220 and errors connected to the determination of the air volume in the chamber) are judged to be at the order of 1 %

The combined uncertainty $U_c\{J_f\}$ of a J_f-determination is found by quadrature summation of the 5 % uncertainty of the radon instrument and the value for $u\{J_f\}$ just discussed.

Open-chamber method (Method A2)

In the open-chamber method, the free exhalation rate is calculated from a modified version of equation 14):

$$J_{f,OC} = (c_{cond} - c_{gas})Q \tag{25}$$

where c_{cond} is the average radon concentration in the chamber from 4 hours after start of conditioning to the time the chamber is closed. The first 4 hours are excluded from the analysis because initially the chamber is filled with room air. c_{gas} is the radon concentration of the nitrogen gas source. In this investigation, c_{gas} was set to zero because a set of four newly purchased cylinders of nitrogen was found to have radon concentrations below 0.05 $Bq\,m^{-3}$. In most cases, gas cylinders were stored for days or weeks before use in the experiment. However, with the exception just given, there was no systematic control of the specific radon concentration of the carrier gas in the actual experiments, and therefore this source of error may bias some of the $J_{f,OC}$-results.

Uncertainty analysis

The statistical variability $u\{J_{f,OC}\}$ of any $J_{f,OC}$-determination is found by quadrature summation of the following contributions:

- The statistical error associated with the measurement of the radon concentration. This source is large as the concentrations are close to zero.

[3] V equals the chamber volume (55.76 L) minus the dead volume of the radon monitor (1.55 L) and the geometric volume of the sample. For a 30 x 30 x 5 cm^3 sample of concrete (4.5 L), V equals $55.76 - 1.55 - 4.5 = 49.71\ L$.

[4] Observe, that a change of λ_{eff} will cause a change of both the fitted estimate of c_∞ and the calculation of J_f as given in equation 24. For example, a typical exhalation rate measurement, a change of 5 % of λ_{eff} will lead to a change of only about 2 % on the final result (i.e. J_f).

- The flow rate Q is judged to be subject to an uncertainty (expressed as one relative standard deviation) of about 10 %. This source of variability result from imperfections of the (manual) flow control (e.g. changes of gas cylinders during experiments).

The possible influence of large c_{gas}-values is not included in the uncertainty estimate.

4.3 Radium-226 measurements

Radium-226 concentration determinations were conducted by Danish Institute for Radiation Hygiene.

5 Experimental results

16 exhalation rate measurements were conducted (identification numbers 103, 105...117, 120 and 121). M7 was measured 7 times, whereas the other 9 materials were measured only once. The measurements were conducted during the period October 22, 1997 to August 10, 1998. Appendix B contains measurement sheets for all measurements.

Figure 3 shows the radon concentration in the chamber during a typical experiment. Initially, the sample is conditioned with a flow of about 1 L min^{-1} for 1 day. Observe, that the concentration has a non-zero value. This is used in the so-called open-chamber method. Then at day 0, the chamber is closed, and the radon concentration increases towards some equilibrium value. This part of the curve is used in the closed-chamber method.

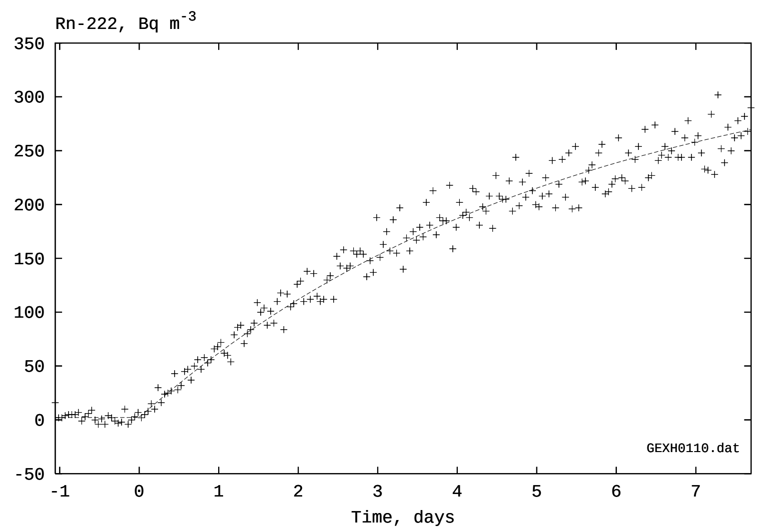

Figure 3. Typical build-up curve. This is measurement no. 110. Day zero on the x-axis is January 21, 1998. The fitted curve: $\hat{c}(t) = c_0 + c_\infty(1 - \exp(-\lambda t))$ has the parameters c_0=2.2±0.9 Bq m^{-3}, and c_∞=350±4 Bq m^{-3}. The free mass-specific exhalation rate is calculated to be $J_{\mathrm{M,f}}$=2.60±0.07 atoms s^{-1} kg^{-1}. The indicated uncertainties are expressed as standard deviations of the given results and include all (known) sources of errors except bias of the radon instrument.

Table 2. Main results obtained with the closed-chamber method. The indicated uncertainties include all (known) sources of error except the uncertainty of the calibration of the radon monitor. All uncertainties are expressed as one standard deviation of the given results. Lightweight aggregate concrete and autoclaved aerated concrete are abbreviated as LAC and AAC, respectively. The fraction of exhalation (in the last column) is the quantity h defined page 4.

ID	Description	Exhalation rate				Radium	Fraction
		Meas.	$J_{A,f} \pm u\{J_{A,f}\}$	$J_{M,f} \pm u\{J_{M,f}\}$		A_{Ra}	of exhal-
		no.	$\mathrm{Bq\,h^{-1}\,m^{-2}}$	$\mathrm{mBq\,h^{-1}\,kg^{-1}}$	$\mathrm{atoms\,s^{-1}\,kg^{-1}}$	$\mathrm{Bq\,kg^{-1}}$	ation, %
M1	LAC, density 600 $\mathrm{kg\,m^{-3}}$	112	0.239±0.009	19.8±0.7	2.62±0.10	32.6[a]	8
M2	LAC type 1, density 1500 $\mathrm{kg\,m^{-3}}$	103	0.584±0.015	19.4±0.5	2.57±0.07	11.8[a]	22
M3	LAC type 2, density 1500 $\mathrm{kg\,m^{-3}}$	110	0.580±0.016	19.6±0.5	2.60±0.07	17.6[a]	15
M4	AAC, density 450 $\mathrm{kg\,m^{-3}}$	111	0.087±0.006	9.5±0.6	1.26±0.08	10.1[b]	12
M5	AAC, density 650 $\mathrm{kg\,m^{-3}}$	109	0.103±0.006	8.0±0.5	1.06±0.06	16.6[a]	6
M6	AAC, density 735 $\mathrm{kg\,m^{-3}}$	108	0.286±0.010	18.9±0.7	2.50±0.09	11.9[a]	21
M7	Ordinary concrete, density 2300 $\mathrm{kg\,m^{-3}}$	105	1.012±0.026	23.9±0.6	3.17±0.08	13.8[a]	23
		114	0.861±0.024	20.4±0.6	2.70±0.07	-	20
		115	0.849±0.021	20.2±0.5	2.67±0.07	-	19
		116	0.859±0.022	20.4±0.5	2.70±0.07	-	20
		117	0.834±0.022	19.8±0.5	2.62±0.07	-	19
		120	0.808±0.021	19.2±0.5	2.54±0.07	-	18
		121	0.814±0.021	19.3±0.5	2.56±0.06	-	19
M8	Gypsum board, density 710 $\mathrm{kg\,m^{-3}}$	106	0.010±0.001	2.4±0.3	0.32±0.04	2.1[c]	15
M9	Bricks, density 1800 $\mathrm{kg\,m^{-3}}$	107	0.020±0.003	0.8±0.1	0.10±0.01	39.8[a]	0.26
M10	Lightweight expanded clay aggregate, density 290 $\mathrm{kg\,m^{-3}}$	113	0.001±0.004	0.1±0.7	0.02±0.10	32.5[b]	0.05

Relative standard uncertainties: [a] 5 %, [b] 10 %, [c] 15 %.

Closed-chamber method

The main results obtained with the closed-chamber method are given in Table 2 together with the results of radium-226 measurements provided by the Danish Institute for Radiation Hygiene. It is seen from the table that concrete-based materials (M1 to M7) have mass-specific exhalation rates in the range from about 1 to 3 $\mathrm{atoms\,s^{-1}\,kg^{-1}}$. The other materials have values below 0.3 $\mathrm{atoms\,s^{-1}\,kg^{-1}}$. Radium-226 concentrations are in the range from about 2 $\mathrm{Bq\,kg^{-1}}$ for gypsum (M8) to 40 $\mathrm{Bq\,kg^{-1}}$ for bricks (M9). Also the fraction of exhalation, h (see definition page 5) varies over a wide range. The lowest value is for M10, where less than 0.1 % of the radon atoms generated in the material escapes the material. The largest fraction of exhalation is for the concrete-based samples M2, M6 and M7, where h amounts to 20 %.

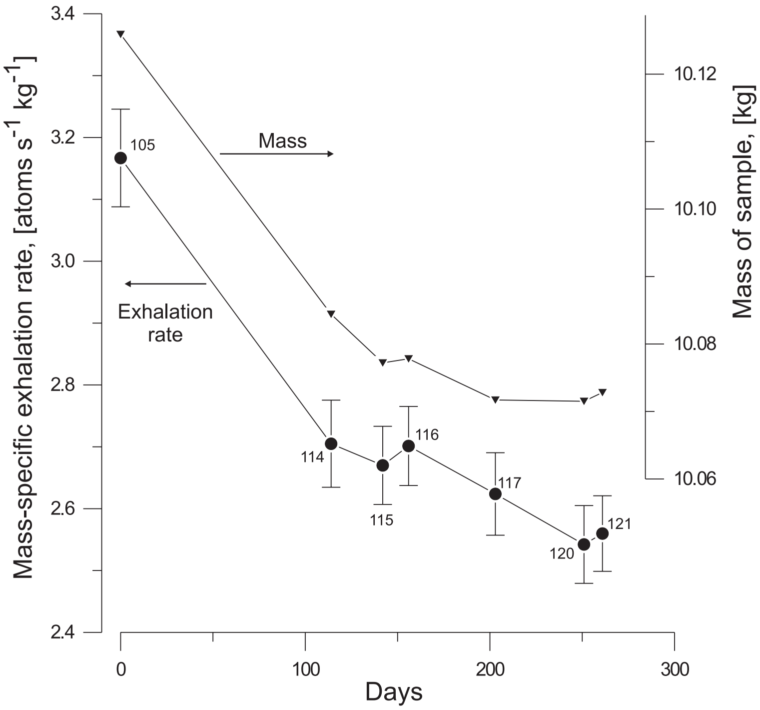

Figure 4. Results for sample M7: Free mass-specific exhalation rate ($J_{\mathrm{M,f}}$) and sample mass versus the time since November 11, 1997.

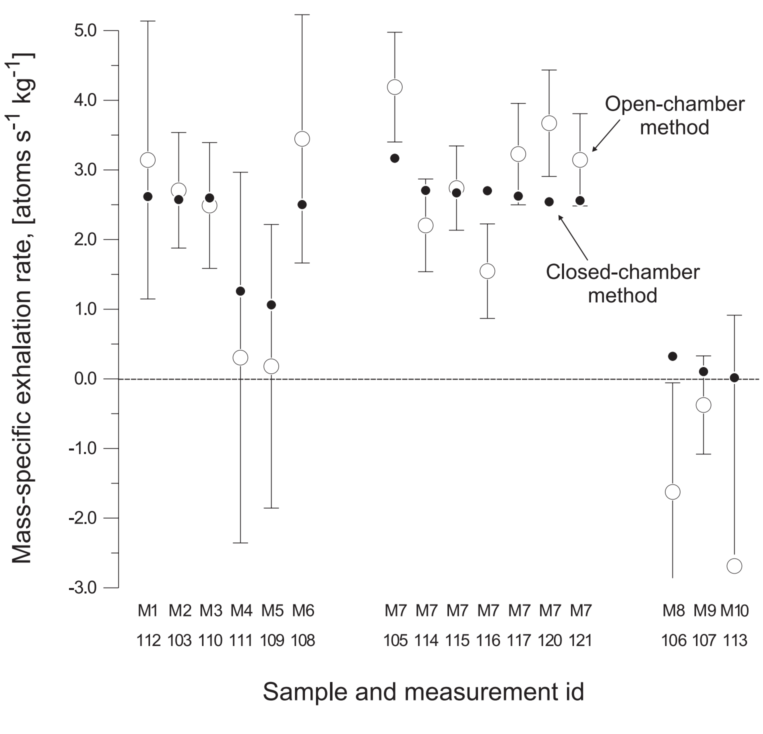

Figure 5. Free mass-specific exhalation rates obtained with the closed-chamber method ($J_{\mathrm{M,f}}$) and the open-chamber method ($J_{\mathrm{M,f,OC}}$). The indicated uncertainties $u\{J_{\mathrm{M,f,OC}}\}$ include all (known) sources of error except the uncertainty of the bias of the radon instrument.

Figure 4 shows the results of 7 (repeated) exhalation rate measurements for sample M7 versus time since the first measurement on November 11, 1997. The total mass of the sample is also shown. As can be seen from the figure, the exhalation rate decreases significantly (i.e. the variability is larger than can be explained by the uncertainty associated with the results). The decrease amounts to about 20 % over a period of 250 days. During the same period, the mass decreases about 0.5 %. The main reason for the change in mass is probably that the samples dries out.

Open-chamber method

The results $J_{\mathrm{M,f,OC}}$ of the open-chamber methods are shown in Figure 5 along with the results for the closed-chamber method $J_{\mathrm{M,f}}$. There is good agreement between the results of the two methods. Observe, that the uncertainty associated with the open-chamber method is large.

Chamber leakage

On five occasions during the period November 7, 1997 to March 20, 1998 special experiments were conducted to test the chamber for leakage. These experiments are presented and discussed page 28.

6 Modelling results

The main cause of exhalation of radon from building materials is believed to be molecular diffusion [St88, Rog94, Re95]. The following investigation therefore focuses on this aspect whereas other mechanisms (for example, exhalation resulting from pressure changes) are ignored. The following issues are investigated:

- Test if an equation of the form $c(t) = c_\infty(1 - \exp(-\lambda t))$ (see page 7 and 11) gives a good description of the radon concentration build-up during an exhalation rate measurement with the closed-chamber method. This is a test of the goodness of the approximation that the exhalation rate J is constant throughout the measurement period.
- Quantify the bias caused by the closed-chamber method (see page 7) compared to an ideal measurement of the free exhalation rate. This is a calculation of the so-called bound-to-free exhalation rate ratio.
- Quantify how sample geometry influences on the measurement result. This aspect is useful for example when results in this investigation are compared with results obtained previously using other sample geometries.
- Quantify how laboratory measurement on small samples (slabs or cylinders) may be extrapolated to full-scale construction parts in real houses.

Although few (if any) published investigations have dealt with the above problems in full 3D and time dependency, many have certainly considered similar situations. The main results can therefore also be extracted from those publications–see for example: [Jo80, Co81, Sa84, Be88, Al94].

Model

All simulations have been performed with the numerical model called `RnMod3d` (see page 10). The model solves the relevant diffusion equation [An92].

Diffusivity, D

It is assumed, that exhalation occurs solely as a result of molecular diffusion, and the main parameter of the hypothetical building material is therefore D, the bulk diffusivity (see page 5). To cover what seems to be the relevant range of diffusivity values, most calculations are performed for values in the range from 10^{-10} m^2 s^{-1} to 10^{-6} m^2 s^{-1}.

Other model parameters

Other parameters of the building materials have (arbitrarily) been set to the following constant values: A_{Ra}=14 Bq kg^{-1} (radium-226 concentration), f=0.2 (fraction of emanation), $\rho_{\mathrm{g}}=2.7 \cdot 10^3$ kg m^{-3} (density of grains), ϵ=0.2 (total porosity), and W=0.0 (water content by mass).

Observe, that with the above values for grain density and porosity, the material has a bulk density of

$$\rho_{\mathrm{m}} = (1-\epsilon)\rho_{\mathrm{g}} = 2.16 \cdot 10^3 \ \mathrm{kg\,m^{-3}} \qquad (26)$$

Also, observe that the emanation rate is:

$$E = f A_{\mathrm{Ra}} = 2.8 \ \mathrm{atoms\,s^{-1}\,kg^{-1}} \qquad (27)$$

The main case under consideration (see next section): a 5 x 30 x 30 cm^3 slab hence has a total mass of M=9.72 kg and a maximum rate of exhalation of $M \cdot E$ = 27.2 atoms s^{-1} or 0.206 Bq h^{-1}.

Model geometry

Several geometries are explored. The main set of calculations concern a slab with dimensions 5 x 30 x 30 cm^3 (4.5 L) placed in a chamber of 58.5 L volume. The free air in the chamber is assumed to be well mixed. Observe, that the free air volume (54.0 L) in the model chamber volume is a bit larger than that of the actual experiment (49.71 L after correction for the rigid volume of the radon monitor, see footnote page 12).

Other geometries that are also considered: cylindric samples and (house) walls with one or two sides facing the indoors. All calculations are performed in full 3D.

Boundary conditions and initial conditions

The chamber is assumed to the leak tight, such that the effective decay constant of the chamber is equal to the decay constant of radon: $\lambda=2.098 \cdot 10^{-6}$ s^{-1}. In some of the calculations the radon concentration of the chamber (or house) is, however, maintained at 0 (free exhalation condition).

Model output

The main output from the model calculations is the total exhalation rate J.

6.1 Simulation of the closed-chamber method

Figure 6 to 8 show simulations of exhalation rate measurements: A sample with dimensions 5 x 30 x 30 cm^3 is placed in a closed chamber initially at zero radon concentration and the radon concentration in the chamber starts to build up. In Figure 6, the sample has a diffusivity D of 10^{-10} m^2 s^{-1}. In Figure 7, D is 10^{-8} m^2 s^{-1}, and in Figure 8, it is as high as 10^{-6} m^2 s^{-1}. The figures show

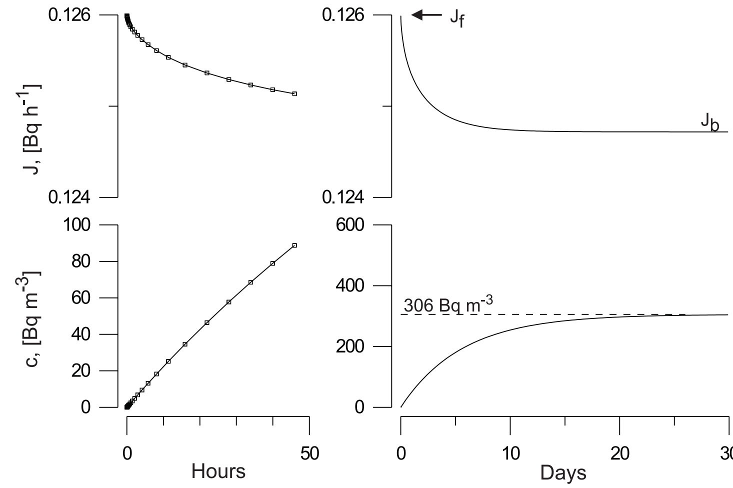

Figure 6. Model simulation of exhalation rate measurement with D=10^{-10} $\mathrm{m^2\ s^{-1}}$.

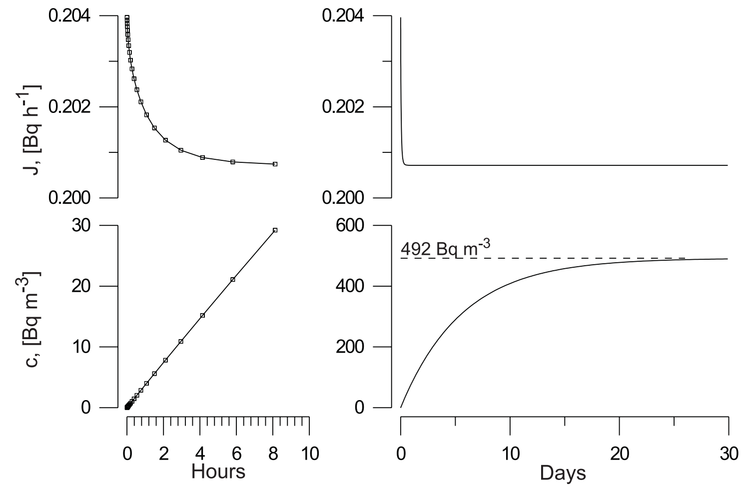

Figure 7. Model simulation of exhalation rate measurement with D=10^{-8} $\mathrm{m^2\ s^{-1}}$.

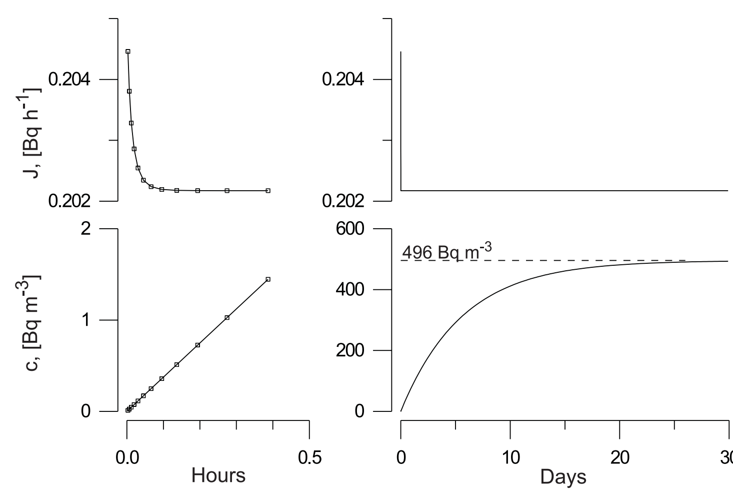

Figure 8. Model simulation of exhalation rate measurement with D=10^{-6} $\mathrm{m^2\ s^{-1}}$.

calculated exhalation rates J as well as calculated radon concentrations c in the chamber. The right plots show the time development over a period of 30 days. The left plots focus on the time immediately after the start of the build-up period.

As marked in the top-right plot of Figure 6, the exhalation rate J is at a maximum at $t = 0$. This is the free exhalation rate J_f. Here, it amounts to $0.126\ \mathrm{Bq\,h^{-1}}$. As time progresses, the exhalation rate decreases towards some equilibrium value J_b–the so-called bound exhalation rate. Here, it amounts to $0.125\ \mathrm{Bq\,h^{-1}}$. Although the plots show a distinctive change in J over time, the relative change is marginal (less than 1 %), and it is a good approximation to treat J as a constant as suggested page 7. As a result of this, the build-up of radon in the chamber $c(t)$ is well described by an equation of the form $c(t) = c_\infty(1 - \exp(-\lambda t))$ (see page 7 and 11).

The results for D=$10^{-8}\ \mathrm{m^2\,s^{-1}}$ (Figure 7) are similar to those just discussed for D=$10^{-10}\ \mathrm{m^2\,s^{-1}}$. Two differences do however exist: the exhalation is much larger and the time it takes to reach the state of bound exhalation is smaller.

For D=$10^{-6}\ \mathrm{m^2\,s^{-1}}$ (Figure 8), the exhalation rate is only slightly higher than that obtained for D=$10^{-8}\ \mathrm{m^2\,s^{-1}}$. This is because these diffusivities are so high that almost all radon will anyway exhale from the sample, regardless of the exact value of D. For D=$10^{-6}\ \mathrm{m^2\,s^{-1}}$ the free exhalation rate J_f amounts to a value just below $0.206\ \mathrm{Bq\,h^{-1}}$ which is the rate radon is produced in the sample (i.e. the maximum exhalation rate–see page 17).

6.2 Bound-to-free exhalation rate ratio

Simulations like the ones just described are conducted for a range of diffusivity values. In each case, the values of the free exhalation rate J_f and the bound exhalation rate J_b are calculated. The bound-to-free exhalation rate ratios:

$$\frac{J_\mathrm{b}}{J_\mathrm{f}} \tag{28}$$

are shown as function of D in Figure 9 for two geometries: a 5 x 30 x 30 $\mathrm{cm^3}$ slab and a 10 x 30 x 30 $\mathrm{cm^3}$ slab.

For the 5 cm slab (i.e. the main geometry used in this study), the bound-to-free exhalation rate ratio is always in the range from about 0.983 to 0.99. Effectively this means that the result obtained with the current closed-chamber method (regardless of the diffusivity of the sample) are biased by less than 2 % (as a result of the effect in question) in comparison with an ideal measurement of J_f. For the measurements with the 10 x 30 x 30 $\mathrm{cm^3}$ slab, the potential bias is a little bit larger. The results are specific for the 58.8 L chamber considered in the calculation.

6.3 g for laboratory samples

The fraction of (mass specific) exhalation-to-emanation g was defined on page 4 as the rate radon exhale from the sample relative to the rate radon is supplied to the pores of the sample (i.e. the rate of emanation, E). Figure 10 shows calculated values of g for a range of sample geometries. The calculations assume equilibrium conditions and that the samples are in an environment with zero radon concentration.

Geometry A is a slab of dimensions 1 x 30 x 30$\mathrm{cm^3}$. The thickness of only 1 cm means that most radon supplied to the pores will escape from the sample regardless of the diffusivity of the material. In contrast, the exhalation rate for a sample formed as a cylinder with 15 cm diameter and 30 cm height will depend somewhat on the diffusivity of the material. For example, imagine a material of diffusivity

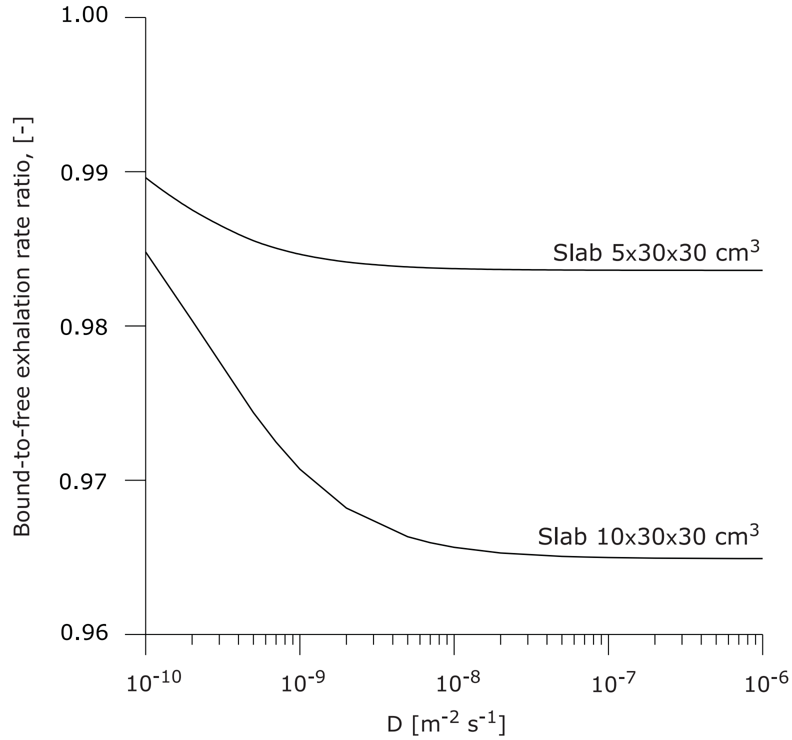

Figure 9. Calculated bound-to-free exhalation rate ratio versus bulk diffusivity for closed-chamber measurements with a 5x30x30 cm^3 slab placed in a 58.5 L chamber.

D=10^{-9} m^2 s^{-1}. Two samples are prepared for exhalation rate measurements: One of geometry A and another of geometry E. Although the materials are the same, Figure 10 shows that the results for geometry E will be about 20 % lower than those obtained with geometry A. For D=10^{-10} m^2 s^{-1}, the difference is about 40 %.

The present investigation mainly applies the 5 x 30 x 30 cm^3 slab geometry (geometry B). It is seen from the figure that even if the diffusivity is a low as 10^{-10} m^2 s^{-1}, this geometry can impede the exhalation only by 30 %.

6.4 g for walls

The fraction of exhalation-to-emanation, g, has been calculated for a range of wall geometries. As shown in Figure 11, both walls having two sides facing the indoors (internal walls) and one side facing the indoors (external walls) are considered. In the one-sided situation, it is assumed that the other side is sealed off, such that no radon can escape from that side.

As can be seen from the figure, there are large differences among the g-values for the different walls. For example, imagine two walls made of the same material: The first wall is 5 cm thick with both sides facing the interior (this is geometry F in Figure 11). The second is a 20 cm thick wall with only one side facing the interior (this is geometry D in Figure 11). We assume that the diffusivity of the material is 10^{-10} m^2 s^{-1}. As can be seen from the figure, g amounts to about 0.56 for the first wall and 0.08 for the second. Hence, as much as 56 % of the radon atoms supplied to the pores of the material degas from the surface of the first wall. The "effectiveness" of degassing is only 8 % for the second wall.

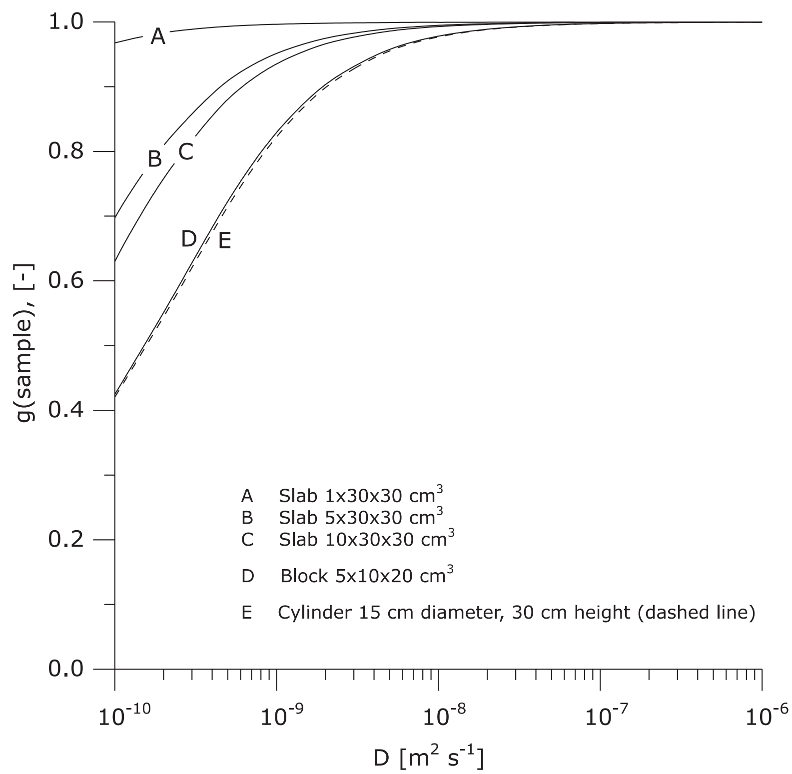

Figure 10. Fraction of exhalation-to-emanation g for a range of sample geometries.

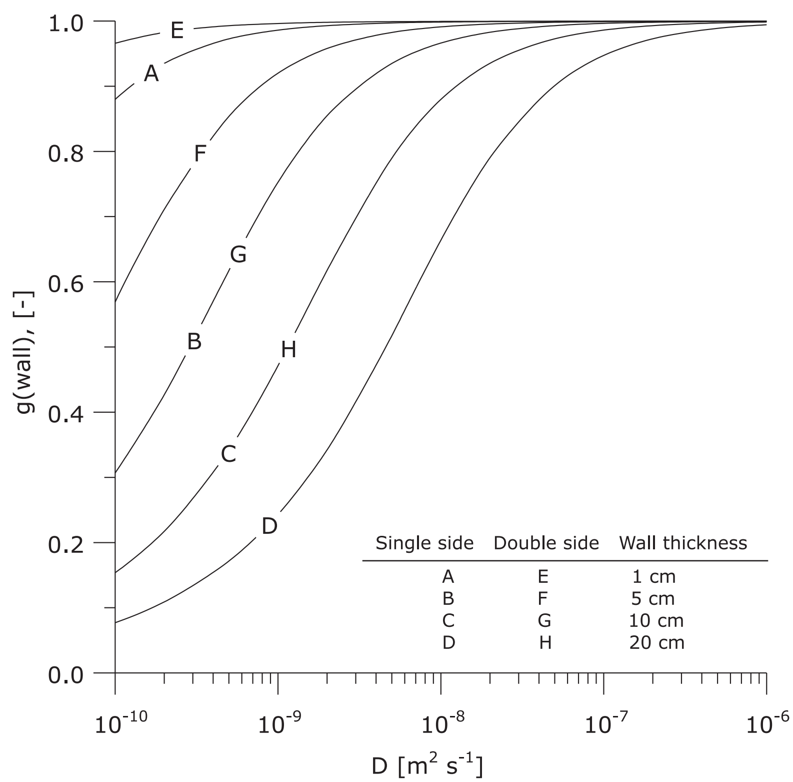

Figure 11. Fraction of exhalation-to-emanation g for different wall geometries.

6.5 From laboratory measurements to full-scale walls

Exact solution

By definition, the exact total exhalation rate $J(\text{wall})$ (Bq h^{-1}) from a wall can be calculated as:

$$J(\text{wall}) = M(\text{wall})\, g(\text{wall})\, E \tag{29}$$

where $M(\text{wall})$ is the mass of the wall, $g(\text{wall})$ is the exhalation-to-emanation ratio for the given wall geometry and diffusivity, and E is the emanation rate of the material. Normally the latter is unknown, and is therefore estimated from laboratory measurement of the mass-specific exhalation rate $J_{\text{M}}(\text{sample})$ of a small sample:

$$E = \frac{J_{\text{M}}(\text{sample})}{g(\text{sample})} \tag{30}$$

such that we obtain:

$$J(\text{wall}) = M(\text{wall})\, \frac{g(\text{wall})}{g(\text{sample})}\, J_{\text{M}}(\text{sample}) \tag{31}$$

which is the formula by which a laboratory measurement can be extrapolated to a full-scale wall. The problem with the extrapolation is, however, that it is only accurate, if the diffusivity of the material is known. In that special case, $g(\text{sample})$ and $g(\text{wall})$ can simply be read from Figure 10 and 11.

Fortunately, two straightforward approximate solutions exist: One based on a high-diffusivity assumption and another based on a low-diffusivity assumption. The true diffusivity of the material will be in between these two limiting cases.

High-diffusivity limit

In the high-diffusivity limit, we assume that the diffusivity of the material is so large that all radon escape both the laboratory sample and the wall (i.e. both are considered to be small compared to the diffusion length of radon). This means that:

$$g(\text{wall}) = 1 \tag{32}$$

$$g(\text{sample}) = 1 \tag{33}$$

such that from equation 31 , we obtain:

$$J(\text{wall}) \approx M(\text{wall})\, J_{\text{M}}(\text{sample}) \tag{34}$$

For real materials, equation 32 and 33 are never fulfilled exactly. However, as long as $g(\text{wall}) \leq g(\text{sample})$, equation 34 provides an overestimate of the wall exhalation rate. Essentially this means that the laboratory sample should be thinner than the wall in question.

Low-diffusivity limit

In the low-diffusivity limit, we assume that the diffusivity is so small that radon escape only from a very thin surface layer of the material. Hence, we assume that the area-specific exhalation rate is the same for the sample and the wall:

$$J_{\text{A}}(\text{wall}) = J_{\text{A}}(\text{sample}) \tag{35}$$

We can therefore write the fractions of exhalation-to-emanation as:

$$g(\text{wall}) = \frac{A(\text{wall})\, J_{\text{A}}(\text{sample})}{M(\text{wall})\, E} \tag{36}$$

$$g(\text{sample}) = \frac{A(\text{sample})\, J_{\text{A}}(\text{sample})}{M(\text{sample})\, E} \tag{37}$$

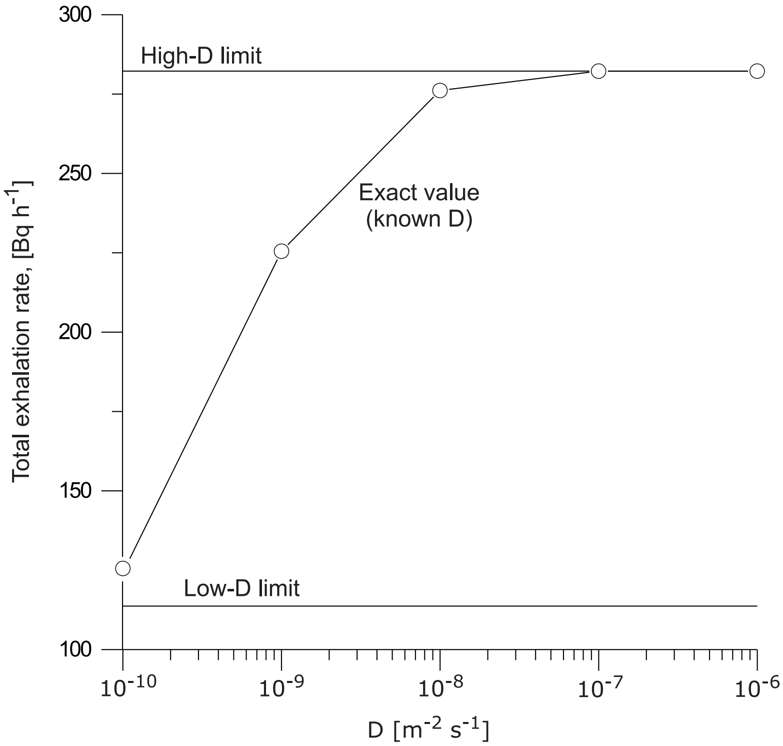

Figure 12. Calculation of the total exhalation rate J(wall) for an internal 10 cm wall. The calculations are based on laboratory measurements of a sample slab of dimensions 5 x 30 x 30 cm^3. *The exact value of J(wall) for each diffusivity D is calculated with equation 31. The horizontal lines are results for the simplified models in equations 34 (the high-diffusivity limit) and equation 38 (the low-diffusivity limit) based on mass-specific and area-specific exhalation rates, respectively.*

Inserting into equation 31 gives:

$$J(\text{wall}) \approx A(\text{wall})\ J_{\text{A}}(\text{sample}) \tag{38}$$

where A(wall) is the surface area of the wall and J_{A}(sample) is the area-specific exhalation rate of the sample. This is an underestimate of the true J(wall) as the diffusivity of real materials is not infinitely low.

Internal wall example

To illustrate how good (or bad) these approximations are we will consider a specific example and compare the approximate solutions with the exact value calculated from equation 31.

Imagine the following situation: Laboratory measurements are conducted on a sample slab of dimensions 5 x 30 x 30 cm^3. Assume, the mass-specific exhalation rate is found to be J_{M}(sample)=19.6 mBq h^{-1} kg^{-1}, and that the area-specific exhalation rate is found to be J_{A}(sample)=0.58 Bq h^{-1} m^{-2}. The material is to be used as an internal house wall of 10 cm thickness. The wall has a mass M(wall) of 14 400 kg, and a total (exposed) surface area of 196 m^2. The question, is what the total exhalation rate will be from the wall? The diffusivity of the material is unknown.

Exact value

First, we will consider the exact (true) value. The 10 cm double sided wall is identified as geometry is G in Figure 11. If the material has a diffusivity D=10^{-9} m^2 s^{-1}, the figure gives g(wall) equal to about 75 %. For the same diffusivity, Figure 10 shows that g(sample) equals 95 %. Hence, from equation 31, the total exhalation rate from the wall $J(\text{wall})$ can be calculated to be: 225 Bq h^{-1}. Likewise, exact results can be obtained for any other hypothetical value of the diffusivity. Results for values in the range from 10^{-10} to 10^{-6} m^2 s^{-1} are shown in Figure 12.

high-D limit Now, in the high-diffusivity limit, the total wall exhalation rate is calculated as the product of the wall mass and the mass-specific exhalation rate of the sample. In the example from above, the estimate of $J(\text{wall})$ amounts to 282 Bq h^{-1}. This value is plotted as the top horizontal line in Figure 12. It is observed, that the approximation is indeed good for high diffusivities.

low-D limit In the low-diffusivity limit, the total wall exhalation rate is calculated as the product of the exposed area of the wall and the area-specific exhalation rate of the sample. This results in the estimate: $J(\text{wall})$=114 Bq h^{-1}. This value is the bottom horizontal line in Figure 12. It is observed, that the approximation is indeed good for low diffusivities.

This example shows that the extrapolation of laboratory measurements to a full wall geometry is subject to considerable uncertainty (a factor of two or such) when the diffusivity of the material is unknown.

7 Reference house calculations

The impact of exhalation from a given building material on the radon concentration in the house where it is used depends on the following factors:

- Mass-specific exhalation rate of the material
- Mass of material in use
- Location of material (e.g. internal wall or external wall)
- House volume
- Air-exchange rate of the house

The building materials investigated in this report have a range of applications. Each application imply that a certain amount of material is used in a certain

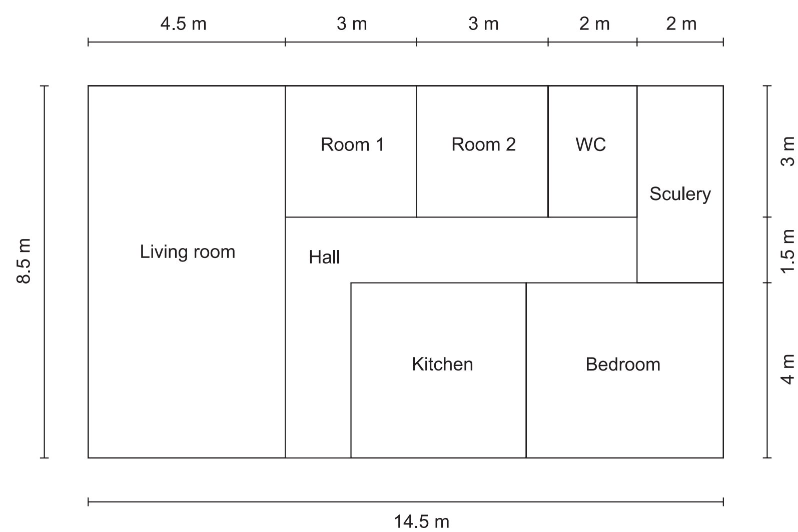

Figure 13. Layout of the reference house used in the assessment of how much the indoor radon concentration increases as a result of specific applications of the investigated building materials.

Table 3. Dimensions of the reference house. The areas of the walls have been corrected for doors (1.8 m² each) and windows (1.4 m² each). The projected area is simply the volume of material in use divided by the thickness (i.e. the cross-sectinal area). The exposed area (A_e) is the area facing the house interior.

Room height	2.5 m
Floor area	123 m^2
House volume (V)	308 m^3
Internal wall with 7 doors	
– Projected area	96 m^2
– Exposed indoor area (A_e)	192 m^2
External wall with 3 doors and 9 windows	
– Projected area	97 m^2
– Exposed indoor area (A_e)	97 m^2
Wall (internal or external) with 10 doors and 9 windows	
– Projected area	193 m^2
– Exposed indoor area (A_e)	289 m^2

part of the house construction. In the following, we assess the impact of these applications for the reference house sketched in Figure 13. The house is a typical Danish slab-on-grade house from the 1970'ies. House dimensions are summarized in Table 3.

Slab-on-grade houese

The diffusivity of the materials has not been measured, and we therefore are bound to estimate exhalation rates in the house on the basis of the approximations described in section 6.5. Those approximations concern the total wall exhalation rate (as if radon from all sides of the wall would exhale into the house), and it is necessary to apply some modifications to account for applications other than internal walls.

Unknown diffusivity

In the *the high-diffusivity limit*, the exhalation rate is calculated from the mass-specific exhalation rate (J_M) as measured in the laboratory for a small sample (see equation 34). We apply a geometry factor G to account for how much of the radon exhaling from the building material that reaches the indoor environment. For contruction parts with one side facing the free atmosphere, G is set to 0.5. For all other locations (internal walls, sub-slab, ceiling construction etc.) we set G to 1. These values for G will probably lead to an overestimation of the exhalation rate. In the *the low-diffusivity limit*, the exhalation rate is calculated from the area-specific exhalation rate (J_A) as measured in the laboratory for a small sample (see equation 38). The wall area is now the exposed area (i.e. the area facing the house interior).

G factor

Exposed area

House concentration

Under steady-state conditions, the rate J ($\mathrm{Bq\,h^{-1}}$) radon is supplied to the house (all sources considered) equals the rate radon is removed:

$$J = \lambda_v V c \tag{39}$$

where λ_v is the air-exchange rate (normally assumed to be 0.5 h^{-1}), V is the volume of the house, and c is the indoor radon concentration ($\mathrm{Bq\,m^{-3}}$). Thus, c can be calculated as:

$$c = \frac{J}{\lambda_v V} \tag{40}$$

In the calculations, we assume that the house air is well mixed, that is has an air exhange og 0.5 h^{-1} and that it can be treated as one single zone.

Building-material applications

In the following the term *external wall* is used to designate a wall that faces the outdoors. In line with this, the term *internal wall* designates a wall that faces only the indoors. The following applications are considered:

Front wall (Danish: *formur*) and back wall (Danish: *bagmur*) are the main components of a cavity wall. The front wall faces the outdoors, and the back wall faces the interior of the house.

Solid load-bearing external wall (Danish: *massiv mur*) An external concrete wall constructed as one single layer.

Internal wall (Danish: *skillevæg*) Walls separating rooms in the house.

Ceiling

Sub-slab layer A layer of large porosity positioned below the slab to prevent ingress of moisture into the floor construction (i.e. a capillary-breaking layer).

Results

Internal wall example

The results of the calculations are shown in Table 4. The top part of the table lists the contribution of each individual material. As an example consider the application of material M1 as a 10 cm internal wall: After correction for doors, the reference house contains internal walls corresponding to an exposed area of 192 m^2 and a mass of 5800 kg. In the laboratory measurements (see Table 2) it was found that M1 has an area-specific exhalation rate (J_A) of 0.239 $Bq\,h^{-1}\,m^{-2}$, and a mass-specific exhalation rate (J_M) of 19.8 $mBq\,h^{-1}\,kg^{-1}$. In the low-diffusivity limit, the internal wall will increase the indoor radon concentration by 0.30 $Bq\,m^{-3}$. This result is given in the second last column of the table. In the high-diffusivity limit, the house concentration will increase by 0.74 $Bq\,m^{-3}$. This is given in the last column of the table. The true contribution of the wall to the radon concentration in the house will be somewhere between 0.30 and 0.74 $Bq\,m^{-3}$.

Full house example

The bottom part of Table 4 gives an example of a full (reference) house constructed with a set of the investigated materials. The calculation shows that the selected building materials could increase the indoor radon by up to 4.3 $Bq\,m^{-3}$. Observe, that the ordinary concrete in the slab accounts for most of the radon. If there is no radon entry from the soil, and if the outdoor air has a radon concentration of 8 $Bq\,m^{-3}$ (as is typical for Denmark), the house will reach a radon concentration of about 12 $Bq\,m^{-3}$ or less.

Air-exchange rate

The results given above apply to the reference house with the air-exchange rate set to 0.5 h^{-1}. Although this value is universally used in such calculations (see e.g. [DS94]), actual measurements [Be94] show that newer Danish houses have an average air-exchange rate of 0.35 h^{-1}. With this air-exchange rate in the reference house, all concentration estimates in Table 4 increase by 43 %. Hence for the full-house example, the contribution of the building materials to the indoor radon concentration would be 6.1 $Bq\,m^{-3}$ or less (and not 4.3 $Bq\,m^{-3}$ or less). Observe, that this value is somewhat less than the value of 11 $Bq\,m^{-3}$ deduced from the measurement of radon levels in multi-family houses given on page 2.

ID	Description	Application	Projected area, m^2	Exposed area, m^2	Thickness mm	Density $kg\,m^{-3}$	Mass 10^3 kg	G	J_A $Bq\,h^{-1}\,m^{-2}$	J_M $mBq\,h^{-1}\,kg^{-1}$	c (from J_A) $Bq\,m^{-3}$	c (from J_M) $Bq\,m^{-3}$
M1	LAC, density 600	Back wall	97	97	100	600	5.8	1	0.239	19.8	0.15	0.75
		Internal wall	96	192	100	600	5.8	1	0.239	19.8	0.30	0.74
M2	LAC type 1, density 1500	Back wall	97	97	100	1500	14.6	1	0.584	19.4	0.37	1.83
		Internal wall	96	192	100	1500	14.4	1	0.584	19.4	0.73	1.81
M3	LAC type 2, density 1500	Back wall	97	97	100	1500	14.6	1	0.58	19.6	0.37	1.85
		Internal wall	96	192	100	1500	14.4	1	0.58	19.6	0.72	1.83
M4	AAC, density 450	Full external wall	97	97	400	450	17.5	0.5	0.087	9.5	0.05	0.54
M5	AAC, density 650	Back wall	97	97	100	650	6.3	1	0.103	8.0	0.06	0.33
		Internal wall	96	192	100	650	6.2	1	0.103	8.0	0.13	0.32
M6	AAC, density 735	Back wall	97	97	100	735	7.1	1	0.286	18.9	0.18	0.87
		Internal wall	96	192	100	735	7.1	1	0.286	18.9	0.36	0.87
M7	Ordinary concrete	Back wall	97	97	100	2300	22.3	1	0.814	19.3	0.51	2.80
		Slab	123	123	100	2300	28.3	1	0.814	19.3	0.65	3.55
		Internal wall	96	192	100	2300	22.1	1	0.814	19.3	1.01	2.77
M8	Gypson board	Ceiling	123	123	13	718	1.1	0.5	0.01	2.4	0.01	0.01
		Walls	289	289	13	718	2.7	1	0.01	2.4	0.02	0.04
M9	Bricks	Front wall	97	97	110	1768	18.9	0.5	0.02	0.8	0.01	0.05
		Back wall	97	97	100	1768	17.1	1	0.02	0.8	0.01	0.09
		Internal wall	96	192	100	1768	17	1	0.02	0.8	0.02	0.09
M10	Lightw. expand. clay agg.	Floor insulation	123	123	280	291	10	1	0.001	0.1	0.00	0.01
Full-house example (see text):												
M7	Ordinary concrete	Slab	123	123	100	2300	28.3	1	0.814	19.3	0.65	3.55
M8	Gypson board	Ceiling	123	123	13	718	1.1	0.5	0.01	2.4	0.01	0.01
M9	Bricks	Front wall	97	97	110	1768	18.9	0.5	0.02	0.8	0.01	0.05
M5	AAC, density 650	Back wall	97	97	100	650	6.3	1	0.103	8.0	0.06	0.33
M5	AAC, density 650	Internal wall	96	192	100	650	6.2	1	0.103	8.0	0.13	0.32
M10	Lightw. expand. clay agg.	Floor insulation	123	123	280	291	10	1	0.001	0.1	0.00	0.01
										Total (full-house example):	0.86	4.27

Table 4. Calculated impact of specific applications of the materials M1 to M10 in the reference house. In the bottom part of the table, a full-house example is shown. Lightweight aggregate concrete and autoclaved aerated concrete are abbreviated as LAC and AAC, respectively.

8 Discussion

8.1 Chamber leakage and other sources of error

A critical assumption linked to the closed-chamber method is that there is no leakage from the chamber. Some efforts was therefore devoted to check this particular aspect of the measurements.

Radon tests

Five radon tests were conducted to identify potential leakage. In each test, a relatively large radon activity was injected into the chamber, and the decay was followed over time. The results are shown in Figure 14. Part (A) of the figure suggests that the decrease of the radon concentration in the chamber is well described by an exponential decay function (i.e. the concentration vs. time curves are linear in a semi-log coordinate system). Part (B) of the figure shows the estimated decay constants normalized with the (true) decay constant for radon (λ=$2.09838 \cdot 10^{-6}$ s^{-1}). Values larger than 1 indicate that radon is removed from the chamber at a rate faster than can be accounted for by radioactive decay. The indicated uncertainties are one standard deviation of the fitted slopes. Experiment (3) indicates a normalized decay constant which is about 9 % larger than unity. The other experiments either are more uncertain or give a normalized decay constant lower than or equal to unity. The weighted mean of the five results is: 0.9965 ± 0.0039. This value is not significantly different from unity.

Pressure tests and one additional radon test

Leaky chamber

Although the radon tests suggest that there is no significant leakage from the chamber, the measured absolute pressure in the chamber (see measurement sheets in Appendix B) reveals that the pressure in the chamber does not remain constant when the chamber is closed! Unfortunately, this was first realized after completion of most of the measurements. A pressure test performed on August 3, 1998 demonstrated that the chamber could not maintain any reasonable room-to-chamber pressure difference for periods much longer than about 10 minutes. The cause of the trouble was found to be a non-standard cable plug mounted on the chamber lid installed in order maintain communication between the continuous radon monitor and a computer. The exhalation rate measurements identified as number 103 to 120 had been conducted with the leaky plug! The final measurement (no. 120) was for material M7. That measurement ended July 28, 1998. The plug was removed on July 29, 1998, and the chamber was pressurized to about 1500 hPa (i.e. to about 500 hPa above the atmospheric pressure). Over a period of 24 h, it was not possible to measure any change in the pressure in the chamber. Sample M7 was then measured one final time. This is measurement no. 121. The difference between the results for measurement 120 and 121 is less than 1 %. This is insignificant when measurement errors are considered.

Correction for leakage

In each individual exhalation-rate measurement, it is (in principle) possible to test if the chamber is subject to leakage: If the effective decay constant is much different from the fixed value assumed in the analysis, it will not be possible to make the theoretical curve fit the experimental data, and the reduced-χ^2 (see equation 23) will reach a value which is statistically different from 1.0. Such a

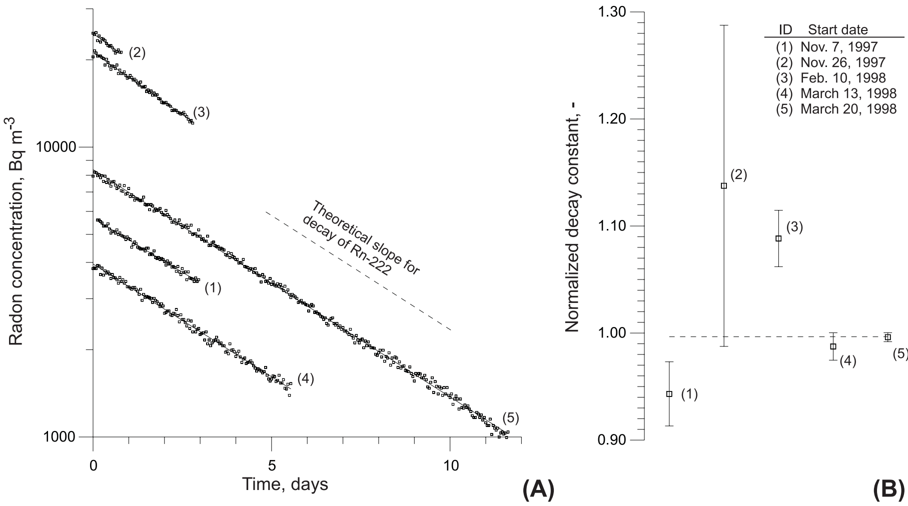

Figure 14. Results of five tests of the tightness of the chamber. Slopes steeper than that indicated in part (A) by the dashed line indicate that radon is removed from the chamber faster than can be explained by radioactive decay. Normalized decay constants are shown in part (B).

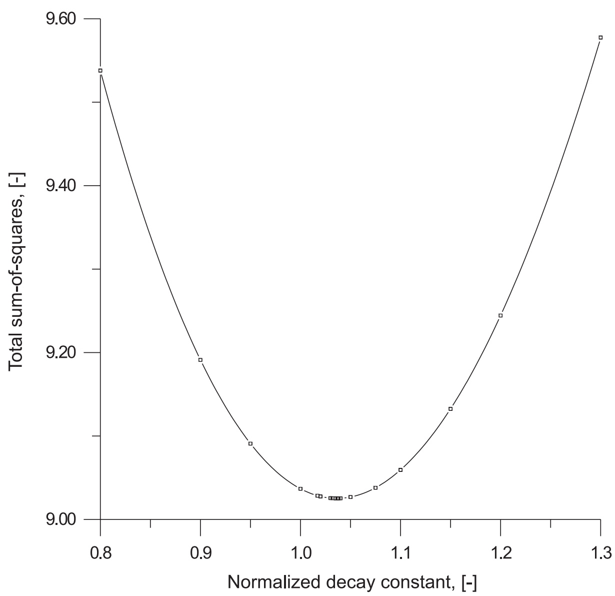

Figure 15. Plot of the sum of all values of the reduced-χ^2 obtained in the experiments number 103 to 120 for a range of (fixed) effective decay constants $\lambda_{\text{eff}}/\lambda$.

test, however, requires that the errors of the radon-concentration measurements are known accurately, which is not the case in this investigation. Here, the results are simply used to find the average leakage during the closed-chamber experiment, such that the results can be corrected for the problem by use of an effective decay constant as described page 7 and 12. Figure 15 shows (grand) sums of the reduced-χ^2 values for the 15 exhalation rate results obtained with the leaky plug (measurement numbers 103 to 120). Each sum is calculated for a fixed value of the effective decay constant λ_{eff}. The value that best describes the (average) leakage present during the experiments is where the curve has its minimum. The value amounts to $\lambda_{\text{eff}} = 1.037 \cdot \lambda$ which is therefore used in the calculation of J_{f} (see equation 24, page 12).

Constant J assumption

Another important assumption behind the closed-chamber method is that the exhalation rate remains constant during the build-up period (see page 7). This problem was addressed by numerical modelling (see page 19). For conditions similar to those of the present experimental set up, it was found that the exhalation rate can be considered to be constant (within about 1.5 %) for materials with diffusivities D in the range from 10^{-10} m^2 s^{-1}to 10^{-6} m^2 s^{-1}. This range of diffusivity values probably covers all materials of interest in this context.

Bound-to-free exhalation ratio

The numerical model was also used to assess the decrease of the exhalation rate as the radon concentration increases in the chamber during the build-up period (see page 19). Calculation of so-called bound-to-free exhalation rate ratios showed that the bound exhalation rate could not be biased (for this reason) by more than about 2 %. A small correction factor α equal to 1.013 was introduced in the calculation of the final result to correct for this problem. See equation 24, page 12.

Comparison with open-chamber method

The best test of bias of a particular measurement procedure is probably to compare its results with results obtained by other means. In this investigation, results of the closed-chamber method can be compared with results of the open-chamber method. The comparison is shown graphically in Figure 5, page 15. It can be seen from the figure that the open-chamber method is subject to considerable uncertainty, but that there seems to be no significant difference between the results obtained with the two methods. The seven measurements of sample M7 are of special interest. The mean of the open-chamber results is 2.96 atoms s^{-1} kg^{-1}, whereas the mean for the closed-chamber method is about 10 % lower: 2.71 atoms s^{-1} kg^{-1}. A t-test of paired samples shows that the difference among results obtained with the two methods is insignificant ($p = 46$ %). This suggests that the closed-chamber method is not strongly biased for example, as a result of chamber leakage or other sources of errors.

Influence of the age of concrete and moisture

It is known from previous investigations [Ul84, St88, Di91], that the exhalation rate of concrete may change over time. For example, Roeloft and Scholten [Roe94] found the exhalation rate of their concrete samples to vary by as much as a factor of 1.5 during the first 6 to 12 months after pouring of the samples. 1 year after pouring, the variability was much less. During the following 6 to 8 years, the exhalation rate decreased monotonously to 0.3–0.6 of the maximum value. In addition, van Dijk and de Jong [Di91] have shown that gypsum as well as concrete samples conditioned to different moisture contents have different exhalation rates. The reason for these observations could be that the emanation rate change with moisture. Changes in diffusivity (for example, as concrete degrade with time) could also be part of the explanation [Rog94]. Furthermore, it has been speculated if vapor transport or change in adsorption characteristics could play a role.

For the above reasons, a 24 h conditioning period at room-like values of temperature and humidity was adopted as part of the measurement protocol. The influence of this particular protocol (or deviations from this[5]) on the final measurement results was not investigated directly. Only the problem of ageing of concrete was studied: Sample M7 was measured 7 times over a period of 250 days (using the same protocol each time). The results in Figure 4 on page 15 show that the exhalation rate decreases by 20 % over the 250 days. The decreases correlates with the decrease in mass. This probably means that the exhalation rate decreases as the sample dries out.

Extrapolation of laboratory results to full walls

As demonstrated by the example page 23, the extrapolation of laboratory exhalation rate measurements to full walls is subject to considerable uncertainty–when the diffusivity of the material is unknown. The true result, however, is bound to be within the so-called low- and high-diffusivity limits. To make conservative estimates, it is best to use the high-diffusivity limit, where the exhalation rate is estimated as the mass of the wall multiplied by the mass-specific radon exhalation rate of the material as measured in the laboratory (see equation 34).

Selected methodology

The main measurements reported here are based on the closed-chamber method identified as Method B on page 7. This method was selected because previous exhalation rate measurements in Denmark had been carried out with this method (see the following section). It also played a role that the instruments needed for the method were readily available at Risø. This report demonstrates that the sources of errors related to the aging of concrete and to the extrapolation of results from laboratory measurements to full-scale houses are generally much larger than the uncertainty associated with the individual laboratory measurements. From this perspective there is probably little need to develop more accurate measurement procedures than the closed-chamber method presented in this work. From the perspective of harmonizing (standard) methods for radon exhalation rate measurements it may, however, be of interest to adopt an open-chamber method equivalent to Method A1 described page 6. This would be in better line with the ongoing standardization work in the Netherlands and Denmark (see page 7).

8.2 Comparison with previous measurements

A number of investigations of radon exhalation rates and radioactivity of Danish building materials were carried out by Jonassen, Ulbak and co-workers in the 1970'ies and 1980'ies. The key references are:

Ulbak, 1980 [Ul80] In 1980, a survey of radioactivity in Danish building materials was carried out by the Danish Institute for Radiation Hygiene [Ul80]. The survey included gamma measurements of potassium-40, thorium-232 and radium-226, but none of radon exhalation rates. As radium-226 is the source of radon, those results can, however, be used to identify candidates for building materials with the highest radon exhalation rates. The emphasis of the

[5] Observe that with the adopted protocol there is no control of humidity in the chamber during the build-up period: if the sample is not in moisture equilibrium with the chamber air when the chamber is closed, then the humidity will change in time. This on the other hand can be seen as a feature of the method: Such deviations from equilibrium are easy to detect with this method.

survey was on bricks and concrete aggregates such as sand, stones and granite. 257 samples were investigated. Few measurements were conducted on real concrete samples. The main results for radium-226 are reproduced in Table 5. Samples believed to be representative for Denmark are marked in the table. The highest radium values were found for aerated alum-shale concrete (of Swedish origin), fly ash, tiles (from various countries), concrete aggregates of granite, and bricks from mo-clay (Danish: *moler*) from Mors. Comparison of Table 2 and 5 shows that there is little difference between the results of the present investigation and the 1980 survey. For example, the radium concentration of brick sample (material M9) in the present investigation is virtually identical to the national average value found in the 1980 survey.

Jonassen and McLaughlin (1976,1980) [**Jo76, Jo80**] reported radon exhalation rates for 14 building material samples. The results for those of the samples believed to have a Danish origin are reproduced in Table 6. Jonassen and McLaughlin used a closed-chamber method for the investigation. The main difference between their procedure and the present work probably is that they used larger chambers (120 L and 200 L) and larger samples (typically 50–100 kg slabs). The linear dimensions of samples are unclear, but the volume-to-surface ratios are given: samples of concrete had volume-to-surface ratios of about 30 m^{-1} which is smaller than the values of about 53 m^{-1} in this investigation (see Table 1, page 9). This probably means that the samples used by Jonassen and McLaughlin were a good deal thicker than the 5 cm used here. This can be of importance for comparison of the results as shown in Figure 10, page 21. The fraction of the chamber volume taken up by the sample ranged from 10 to 55 % with a typical value around 40 %. The chamber used in this investigation is in all cases filled less than 10 %. It is unclear how the samples were conditioned in the chamber.

The values reported by Jonassen and McLaughlin and those found in this investigation are surprisingly similar. For example, the values for mass-specific exhalation (J_{M}) for ordinary concrete was found by Jonassen and McLaughlin to be about 2.0–2.4 atoms $s^{-1}\,kg^{-1}$ (sample J1 and J2) compared to 2.6 atoms $s^{-1}\,kg^{-1}$ found here for sample M7. The mass-specific exhalation rate for bricks (solid type) of 0.08 atoms $s^{-1}\,kg^{-1}$ (J11) is identical to the value of 0.10 atoms $s^{-1}\,kg^{-1}$ found here for sample M9. It is probably reasonable to compare the lightweight concrete samples J7 (density 750 $kg\,m^{-3}$) and J8 (density 780 $kg\,m^{-3}$) with sample M1 (density 600 $kg\,m^{-3}$) of the present investigation. It is seen from the tables, that the area-specific exhalation rates are almost identical (about 0.2 $Bq\,h^{-1}\,m^{-2}$ in both cases). The mass-specific exhalation rates, do however, deviate by a factor of 1.9: J7 has a mass-specific exhalation rate of 1.4 atoms $s^{-1}\,kg^{-1}$, while the value of M1 is 2.6 atoms $s^{-1}\,kg^{-1}$. The reason the area-specific exhalation rates agree while the mass-specific values do no, could be that radon exhale only from a relatively thin surface layer of the sample (corresponding to the low-diffusivity limit discussed page 23). The larger surface-to-volume ratios of the samples used by Jonassen and McLaughlin will produce lower mass-specific exhalation rates compared with this investigation. This is supported by the observation that M1 has a fraction of exhalation of only 8 %.

Ulbak, Jonassen and Bækmark (1984) [**Ul84**] investigated radon exhalation rates for cylindrical concrete samples (15 cm in diameter and 30 cm in height). This is geometry E in Figure 10. Among other things, it was found:

- that radon exhalation rate measurements conducted during the first year after production were very variable. In the present investigation, no such variability was observed. It was, however, observed that the exhalation

Table 5. Selected radium-226 results (A_{Ra}) from the 1980-survey of radioactivity in Danish building materials reported by Ulbak [Ul80]. The results are sorted in order of decreasing average radium concentration.

Group	Number of samples	A_{Ra} ($\mathrm{Bq\,kg^{-1}}$) min	average	max
Alum-shale aerated concrete	2		670	
Fly ash[a]	10	110	150	210
Tiles from various counties	13	22	66	108
Bricks[a]	79	23	42	86
Leca	3	36	40	43
Concrete aggregates[a] (e.g. sand, stones, granite)	107	< 4	19	95
Concrete	6	13	16	24
Aerated concrete	3	9	15	25
Natural gypsum[a]	12	< 4	8	13

[a] Representative value for Denmark.

Table 6. Results of Danish exhalation rate measurements as reported by Jonassen and McLaughlin [Jo76, Jo80].

ID	Description	Exhalation rate J_A ($\mathrm{Bq\,h^{-1}\,m^{-2}}$)	J_M ($\mathrm{mBq\,h^{-1}\,kg^{-1}}$)	J_M ($\mathrm{atoms\,s^{-1}\,kg^{-1}}$)
J1	Ordinary concrete, gravel and sand from the sea, Danish deposits, density 2300 $\mathrm{kg\,m^{-3}}$	1.3	18	2.38
J2	Ordinary concrete, gravel and sand from pits, Danish deposits, density 2200 $\mathrm{kg\,m^{-3}}$	1.0	15	1.98
J7	Lightweight concrete, Danish origin, clay based, density 750 $\mathrm{kg\,m^{-3}}$	0.24	11	1.42
J8	Lightweight concrete, Danish origin, clay based, density 780 $\mathrm{kg\,m^{-3}}$	0.16	11	1.42
J9	Expanded clay concrete, Leca, density 650 $\mathrm{kg\,m^{-3}}$	0.16	9	1.19
J10	Bricks, solid type, density 1900 $\mathrm{kg\,m^{-3}}$	0.017	0.6	0.08
J11	Bricks, cavity type, density 1900 $\mathrm{kg\,m^{-3}}$	0.007	0.6	0.08
J14	Gypsum board, density 980 $\mathrm{kg\,m^{-3}}$	0.005	0.8	0.11

rate of sample M7 changed over time. For a 250 day period, the exhalation rate decreased 20 %.

- that the use of fly ash as a substitute for cement in ordinary concrete had no significant impact on the radon exhalation rates (when compared with ordinary concretes).
- and that ordinary Danish concrete has a mass-specific exhalation rate of 17 $\mathrm{mBq\,h^{-1}\,kg^{-1}}$ (i.e. 2.25 $\mathrm{atoms\,s^{-1}\,kg^{-1}}$). This is only slightly less than the value of about 20 $\mathrm{mBq\,h^{-1}\,kg^{-1}}$ found in this investigation for sample M7. This result is interesting as the ordinary concrete studied

by Ulbak et al. has a radium-226 contents of about 34 $Bq\,kg^{-1}$ compared with the value of only 13.8 $Bq\,kg^{-1}$ for sample M7. This means that radon exhales more "efficiently" from M7 compared to the concrete studied by Ulbak et al. In other words, the fraction of exhalation h are different: 6.6 % (= 2.25/34) versus about 20 % for sample M7. The difference could be related to differences in radium content and transport properties of the aggregates used for the two concretes. Also part of the explanation could be measurement errors related to the geometry of the samples. From Figure 10, it can be seen that the cylindrical geometry used by Ulbak et al. (for any given diffusivity) impedes the radon exhalation more than the 5 x 30 x 30 cm^3 geometry used in the present study. If the diffusivity of both materials happens to be as low as 10^{-10} $m^2\,s^{-1}$, the slab should have an exhalation rate about 0.70/0.42 = 67 % larger than that of the cylinder (g(sample) = 0.70 for the slab and 0.42 for the cylinder, see Figure 10).

9 Conclusions

A procedure for measurement of radon exhalation rates from building material samples has been established at Risø National Laboratory, and the method has been applied to 10 samples supplied by H+H Industri A/S, Ølsted, Denmark. The applied closed-chamber method is similar to the one previously used in Denmark by Jonassen and co-workers.

With respect to the analytical aspects of the method, the following conclusions were reached :

- The applied closed-chamber method was found to be in need of minor corrections relating to leakage and the problem of bound exhalation. With these corrections, it was not possible to demonstrate any significant difference between the results obtained with the closed-chamber method and an open-chamber method. Since these two methods are sensitive to different types of errors this suggests that the closed-chamber method (as applied in this work) provides results of good quality.
- It was demonstrated that sources of errors related to the aging of concrete and to the extrapolation of results from laboratory measurements to full-scale houses are generally much larger than the assessed accuracy of any of the laboratory measurements. From this perspective, there is probably little need to develop more accurate measurement procedures than the closed-chamber method presented in this work. Only from the perspective of harmonizing (standard) methods for radon exhalation rate measurements it could be of interest to adopt a more accurate open-chamber method than was used in this work.

The following sample specific conclusions were reached:

- The exhalation rates were in the range from about 1 to 3 atoms $s^{-1}\,kg^{-1}$.
- For a typical single-family house, the contribution of any single application of the materials was 3.6 $Bq\,m^{-3}$ or below. A reference house build exclusively from a set of the investigated materials was found to have an indoor concentration of 4.3 $Bq\,m^{-3}$ or below (all other sources neglected). This value is at the same level as the outdoor radon concentration.
- The results reported here are essentially identical to those reported in by Jonnassen, Ulbak and co-workers.

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A Guide to measurement sheets

The following list describes the type of information listed on the measurement sheets (see next appendix):

Method (primary) This line of text tells that the closed-chamber method (method B, page 11) is used as primary method.

Method (secondary) This line of text tells that the measurement sheet also contains data based on the open-chamber method identified as Method A2 (see page 12). Results obtained with this method are relatively uncertain in comparison with the primary method. The open-chamber results are given mainly to help identify gross errors.

Measurement procedure This line of text describes the measurement procedure.

Measurement no. / Series / Sample ID Each measurement has a unique identification number. The series and the sample are identified in free text format.

Sample descriptor Description of the sample.

Chamber / Carrier gas Sample and carrier gas identification.

Radon instrument / Cycle time Identification of the radon monitor in use and the selected cycle time (10 m or 1 h).

Analysis program / Datafile / Graphfile Identification of the data analysis program and data files.

Date of reporting The date the results were reported. If the results have been revised, then it is the date of the last revision.

Experimenter Name of the person who carried out the experimental work.

$J_{\mathrm{M,f}}$ The free mass-specific exhalation rate of the sample. This is the main result of the measurement. It is obtained with the closed-chamber method. The term *free* means that the exhalation rate given corresponds to the exhalation rate of the sample when it is placed in an environment with air maintained at zero radon concentration (see page 3 and 5).

$\mathrm{U_c}\{J_{\mathrm{M,f}}\}$ The combined uncertainty (expressed as one standard deviation) of the measurement result $J_{\mathrm{M,f}}$. All known sources of errors are included.

u$\{J_{\mathrm{M,f}}\}$ This is the same as $\mathrm{U_c}\{J_{\mathrm{M,f}}\}$ except that the uncertainty of the radon monitor bias is not included.

$J_{\mathrm{A,f}}$ The free area-specific exhalation rate of the sample.

$\mathrm{U_c}\{J_{\mathrm{A,f}}\}$ The combined uncertainty (expressed as one standard deviation) of the measurement results $J_{\mathrm{A,f}}$. All known sources of errors are included.

u$\{J_{\mathrm{A,f}}\}$ This is the same as $\mathrm{U_c}\{J_{\mathrm{A,f}}\}$ except that the uncertainty of the radon monitor bias is not included.

$J_{\mathrm{M,f,OC}}$ An estimate of the free mass-specific exhalation rate as found with the open-chamber method. The used open-chamber method is considered to be much less accurate than the closed-chamber method, and the open chamber results are given only to help identify gross errors.

u$\{J_{\mathrm{M,f,OC}}\}$ The combined uncertainty of the measurement results $J_{\mathrm{M,f,OC}}$. All known sources or errors (except the uncertainty of the bias of the radon monitor) are included.

Sample dimensions This gives the overall shape and dimension of the sample.

Mass (before, after and lost) The sample is weighted before it is put in the chamber and after it is taken out (i.e. after completion of the exhalation rate measurement). The mass lost is the difference between the two masses. Typically mass is lost by the drying out of the sample or because sample grains break off.

Volume, Area, Density This is the bulk volume of the sample, the total surface area, and the density of the sample.

Empty chamber vol. etc. This field gives the total volume of the chamber, the volume of rigid things inside the chamber such as the radon monitor (but not the sample), and the air volume in the chamber during measurements after correction for dead space and sample.

Model equation This is the main equation used in the non-linear fitting of the radon concentration in the chamber. The full equation is given as equation 21, page 11.

Chi-2 reduced, N This is a measure of the non-linear fit (see page 11). N is the number of individual measurements involved.

Fitted parameter c_0**, s**$\{c_0\}$ This is the result of fitting the equation to the data. $\mathrm{s}\{c_0\}$ is the standard deviation of the fitted parameter c_0 (sometimes called the standard error).

Fitted parameter c_∞**, s**$\{c_\infty\}$ This is the result of fitting the equation to the data. $\mathrm{s}\{c_\infty\}$ is the standard deviation of the fitted parameter c_∞ (sometimes called the standard error).

Fixed parameter λ_{eff} The effective decay constant is set to a fixed value in the fitting equation. λ_{eff} is expressed as a factor multiplied by the decay constant of radon-222 ($2.09838 \cdot 10^{-6}\ \mathrm{s}^{-1}$). $\lambda_{\mathrm{eff}} > \lambda$ means that the chamber is leaky.

Bound-to-free exhalation correction This factor attempts to convert the measured bound exhalation rate to a free exhalation rate (see page 12).

Start and Stop This gives the dates and times for the conditioning period (first column) and the build-up period (second column).

Period This gives the total duration in days of the conditioning period (first column) and the build-up period (second column).

Q_{wet} This is the setting of the mass-flow controller in the "wet flow line".

$Q{=}Q_{\text{wet}}{+}Q_{\text{dry}}$ This is the total flow rate of air leaving the chamber as measured with the flow meter at the pressure and temperature given under notes (see Figure 2, page 9).

c $\mathbf{s}\{c\}$ This is the mean and standard deviation of the radon concentration measurements in the chamber made during the conditioning period (first column) and the build-up period (second column).

T $\mathbf{s}\{T\}$ This is the mean and standard deviation of the temperature measurements made in the chamber during the conditioning period (first column) and the build-up period (second column).

RH s{RH} This is the mean and standard deviation of the relative humidity measurements made in the chamber during the conditioning period (first column) and the build-up period (second column).

P_{atm} $\mathbf{s}\{P_{\text{atm}}\}$ This is the mean and standard deviation of the pressure measurements made in the chamber during the conditioning period (first column) and the build-up period (second column).

N This is the number of measurements (of c, T, RH, or P_{atm}) made in the chamber during the conditioning period (first column) and the build-up period (second column).

Figures The figures show the radon concentration, pressure, relative humidity, and temperature in the chamber during the conditioning and the build-up period. The chamber is closed at time t=0.

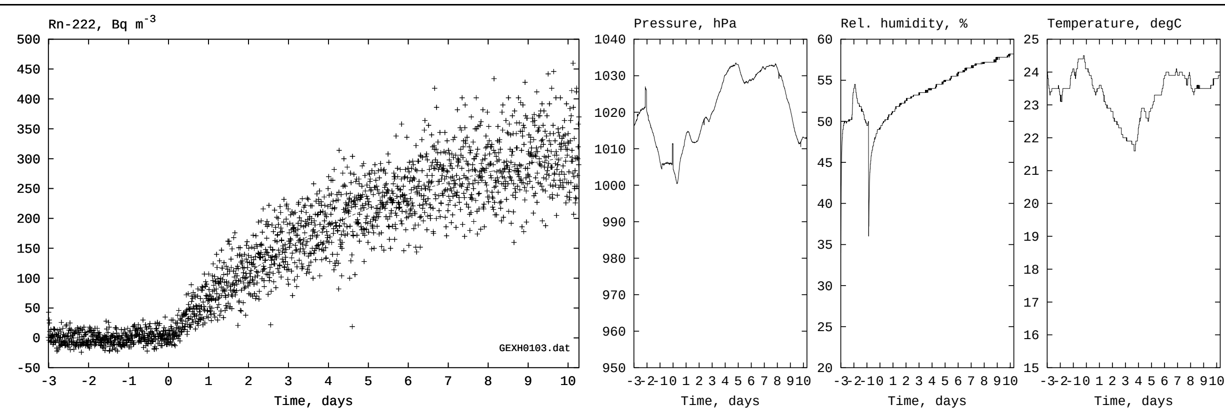
B Measurement sheets

This appendix contains measurement sheets with detailed results for all exhalation rate determinations.

^{222}Rn exhalation rate measurement

Method (primary)	Closed-chamber method w. continuous radon monitor
Method (secondary)	Open-chamber method w. continuous radon monitor
Measurement procedure	Conditioning and build-up (June 1998 procedure)
Measurement no. / Series / Sample ID	#0103 / H+H Industri A/S / M2
Sample descriptor	LAC type 1, density 1500
Chamber / Carrier gas	RnChamber2 / nitrogen
Radon instrument / Cycle time	AlphaGuard PQ2000 EF-231 / 10 min
Analysis program / Datafile / Graphfile	`ExhBas02.pas` / `ExhData4.dat` / `GEXH0103.dat`
Date of reporting	August 20, 1998
Experimenter	Claus E. Andersen, Risø National Lab., Denmark
Free mass-specific exhalation rate $J_{\mathrm{M,f}}$	2.57 atoms $\mathrm{s^{-1}\,kg^{-1}}$
Combined uncertainty ($k=1$) $\mathrm{U_c}\{J_{\mathrm{M,f}}\}$	0.15 atoms $\mathrm{s^{-1}\,kg^{-1}}$
Type A + B uncert. except Rn monitor bias $\mathrm{u}\{J_{\mathrm{M,f}}\}$	0.07 atoms $\mathrm{s^{-1}\,kg^{-1}}$
Free area-specific exhalation rate, $J_{\mathrm{A,f}}$	0.584 Bq $\mathrm{h^{-1}\,m^{-2}}$
Combined uncertainty ($k=1$) $\mathrm{U_c}\{J_{\mathrm{A,f}}\}$	0.034 Bq $\mathrm{h^{-1}\,m^{-2}}$
Type A + B uncert. except Rn monitor bias $\mathrm{u}\{J_{\mathrm{A,f}}\}$	0.015 Bq $\mathrm{h^{-1}\,m^{-2}}$
Open-chamber result $J_{\mathrm{M,f,OC}}$ / $\mathrm{u}\{J_{\mathrm{M,f,OC}}\}$	2.71 atoms $\mathrm{s^{-1}\,kg^{-1}}$/ 0.83 atoms $\mathrm{s^{-1}\,kg^{-1}}$
Sample dimension	Slab 29.8 cm x 30.0 cm x 5.4 cm
Mass	7.3170 kg
Volume (V) / Area (A) / Density (ρ_{m})	4.83 L / 0.243 $\mathrm{m^2}$/ 1516 $\mathrm{kg\,m^{-3}}$
Empty chamber vol. / dead space (−sample) / air	55.76 L / 1.55 L / 49.38 L
Model equation	$c(t) = c_0 + c_\infty(1-\exp(-\lambda_{\mathrm{eff}}t))$
Chi-2 reduced (χ^2_ν) / N	0.775 / 125 + 1478 = 1603
Fitted parameter c_0 / $\mathrm{s}\{c_0\}$	-4.83 Bq $\mathrm{m^{-3}}$ / 0.51 Bq $\mathrm{m^{-3}}$
Fitted parameter c_∞ / $\mathrm{s}\{c_\infty\}$	363.05 Bq $\mathrm{m^{-3}}$ / 2.79 Bq $\mathrm{m^{-3}}$
Fixed parameter λ_{eff}	$1.037\cdot\lambda$, where $\lambda = 2.09838\cdot10^{-6}\ \mathrm{s^{-1}}$
Bound-to-free exhalation correction ($J_{\mathrm{M,f}}/J_{\mathrm{M}}$)	1.013

		Conditioning		Build-up		Unit	Notes
Start			221097-17:20		231097-14:01		
Stop			231097-14:01		021197-20:30		
Period			0.86		10.27	d	
Q_{wet}			500		0	$\mathrm{mL_n\,min^{-1}}$	Mass-flow controlled
Q=Q_{wet}+Q_{dry}			988.9		0	$\mathrm{mL\,min^{-1}}$	Measured at 1005 hPa and 25 °C
c	$\mathrm{s}\{c\}$	3.7	1.0	203.0	2.5	Bq $\mathrm{m^{-3}}$	
T	$\mathrm{s}\{T\}$	24.3	0.0	23.2	0.0	°C	
RH	s{RH}	46.9	0.2	54.8	0.1	%	
P_{atm}	$\mathrm{s}\{P_{\mathrm{atm}}\}$	1006.3	0.1	1022.5	0.2	hPa	
N		125		1478		-	Number of measurements

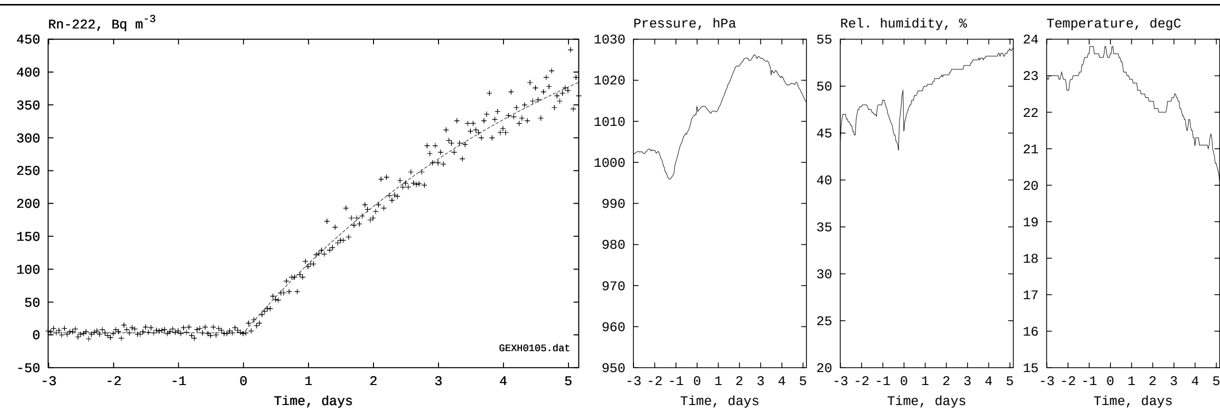


Notes: -

^{222}Rn exhalation rate measurement

Method (primary)	Closed-chamber method w. continuous radon monitor
Method (secondary)	Open-chamber method w. continuous radon monitor
Measurement procedure	Conditioning and build-up (June 1998 procedure)
Measurement no. / Series / Sample ID	#0105 / H+H Industri A/S / M7
Sample descriptor	Ordinary concrete, density 2300
Chamber / Carrier gas	RnChamber2 / nitrogen
Radon instrument / Cycle time	AlphaGuard PQ2000 EF-231 / 1 h
Analysis program / Datafile / Graphfile	`ExhBas02.pas` / `ExhData4.dat` / `GEXH0105.dat`
Date of reporting	August 20, 1998
Experimenter	Claus E. Andersen, Risø National Lab., Denmark
Free mass-specific exhalation rate $J_{\mathrm{M,f}}$	3.17 atoms $\mathrm{s^{-1}\,kg^{-1}}$
Combined uncertainty ($k=1$) $\mathrm{U_c}\{J_{\mathrm{M,f}}\}$	0.18 atoms $\mathrm{s^{-1}\,kg^{-1}}$
Type A + B uncert. except Rn monitor bias $\mathrm{u}\{J_{\mathrm{M,f}}\}$	0.08 atoms $\mathrm{s^{-1}\,kg^{-1}}$
Free area-specific exhalation rate, $J_{\mathrm{A,f}}$	1.012 $\mathrm{Bq\,h^{-1}\,m^{-2}}$
Combined uncertainty ($k=1$) $\mathrm{U_c}\{J_{\mathrm{A,f}}\}$	0.058 $\mathrm{Bq\,h^{-1}\,m^{-2}}$
Type A + B uncert. except Rn monitor bias $\mathrm{u}\{J_{\mathrm{A,f}}\}$	0.026 $\mathrm{Bq\,h^{-1}\,m^{-2}}$
Open-chamber result $J_{\mathrm{M,f,OC}}$ / $\mathrm{u}\{J_{\mathrm{M,f,OC}}\}$	4.19 atoms $\mathrm{s^{-1}\,kg^{-1}}$/ 0.79 atoms $\mathrm{s^{-1}\,kg^{-1}}$
Sample dimension	Slab 29.9 cm x 30.0 cm x 5.0 cm
Mass (before) / Mass (after) / Mass (lost)	10.1264 kg / 10.1253 kg / 1.1 g
Volume (V) / Area (A) / Density (ρ_{m})	4.49 L / 0.239 $\mathrm{m^2}$/ 2258 $\mathrm{kg\,m^{-3}}$
Empty chamber vol. / dead space (−sample) / air	55.76 L / 1.55 L / 49.72 L
Model equation	$c(t) = c_0 + c_\infty(1 - \exp(-\lambda_{\mathrm{eff}}t))$
Chi-2 reduced (χ^2_ν) / N	0.612 / $73 + 124 = 197$
Fitted parameter c_0 / $\mathrm{s}\{c_0\}$	3.19 $\mathrm{Bq\,m^{-3}}$ / 0.61 $\mathrm{Bq\,m^{-3}}$
Fitted parameter c_∞ / $\mathrm{s}\{c_\infty\}$	613.93 $\mathrm{Bq\,m^{-3}}$ / 6.34 $\mathrm{Bq\,m^{-3}}$
Fixed parameter λ_{eff}	$1.037 \cdot \lambda$, where $\lambda = 2.09838 \cdot 10^{-6}\ \mathrm{s^{-1}}$
Bound-to-free exhalation correction ($J_{\mathrm{M,f}}/J_{\mathrm{M}}$)	1.013

		Conditioning		Build-up		Unit	Notes
Start		111197-14:34		141197-16:11			
Stop		141197-16:11		191197-20:18			
Period			3.07		5.17	d	
Q_{wet}			500		0	$\mathrm{mL_n\,min^{-1}}$	Mass-flow controlled
$Q{=}Q_{\mathrm{wet}}{+}Q_{\mathrm{dry}}$			1174.0		0	$\mathrm{mL\,min^{-1}}$	Measured at 1012 hPa and 23 °C
c	$\mathrm{s}\{c\}$	4.6	0.5	226.3	10.2	$\mathrm{Bq\,m^{-3}}$	
T	$\mathrm{s}\{T\}$	23.2	0.0	22.1	0.1	°C	
RH	s{RH}	46.8	0.2	51.5	0.2	%	
P_{atm}	$\mathrm{s}\{P_{\mathrm{atm}}\}$	1003.0	0.5	1019.7	0.4	hPa	
N		73		124		-	Number of measurements

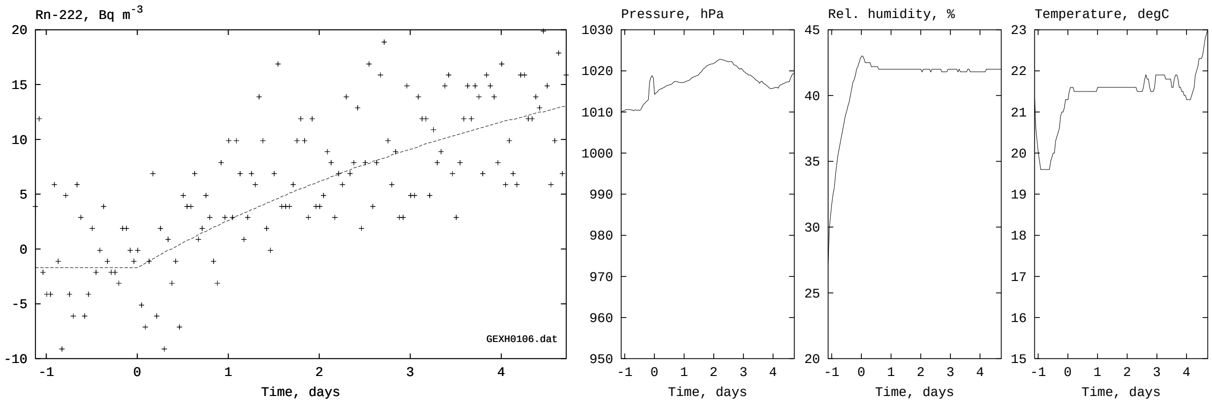


Notes: -

^{222}Rn exhalation rate measurement

Method (primary)	Closed-chamber method w. continuous radon monitor
Method (secondary)	Open-chamber method w. continuous radon monitor
Measurement procedure	Conditioning and build-up (June 1998 procedure)
Measurement no. / Series / Sample ID	#0106 / H+H Industri A/S / M8
Sample descriptor	Gypsum board
Chamber / Carrier gas	RnChamber2 / nitrogen
Radon instrument / Cycle time	AlphaGuard PQ2000 EF-231 / 1 h
Analysis program / Datafile / Graphfile	`ExhBas02.pas` / `ExhData4.dat` / `GEXH0106.dat`
Date of reporting	August 20, 1998
Experimenter	Claus E. Andersen, Risø National Lab., Denmark
Free mass-specific exhalation rate $J_{\mathrm{M,f}}$	0.32 atoms $\mathrm{s^{-1}\,kg^{-1}}$
Combined uncertainty ($k=1$) $\mathrm{U_c}\{J_{\mathrm{M,f}}\}$	0.05 atoms $\mathrm{s^{-1}\,kg^{-1}}$
Type A + B uncert. except Rn monitor bias $\mathrm{u}\{J_{\mathrm{M,f}}\}$	0.04 atoms $\mathrm{s^{-1}\,kg^{-1}}$
Free area-specific exhalation rate, $J_{\mathrm{A,f}}$	0.010 $\mathrm{Bq\,h^{-1}\,m^{-2}}$
Combined uncertainty ($k=1$) $\mathrm{U_c}\{J_{\mathrm{A,f}}\}$	0.001 $\mathrm{Bq\,h^{-1}\,m^{-2}}$
Type A + B uncert. except Rn monitor bias $\mathrm{u}\{J_{\mathrm{A,f}}\}$	0.001 $\mathrm{Bq\,h^{-1}\,m^{-2}}$
Open-chamber result $J_{\mathrm{M,f,OC}}$ / $\mathrm{u}\{J_{\mathrm{M,f,OC}}\}$	-1.62 atoms $\mathrm{s^{-1}\,kg^{-1}}$/ 1.57 atoms $\mathrm{s^{-1}\,kg^{-1}}$
Sample dimension	5 plates of 29.8 cm x 29.8 cm x 1.3 cm
Mass (before) / Mass (after) / Mass (lost)	3.9595 kg / 3.9571 kg / 2.4 g
Volume (V) / Area (A) / Density (ρ_{m})	5.55 L / 0.963 $\mathrm{m^2}$/ 713 $\mathrm{kg\,m^{-3}}$
Empty chamber vol. / dead space ($-$sample) / air	55.76 L / 1.55 L / 48.66 L
Model equation	$c(t) = c_0 + c_\infty(1 - \exp(-\lambda_{\mathrm{eff}} t))$
Chi-2 reduced (χ^2_ν) / N	0.614 / $27 + 114 = 141$
Fitted parameter c_0 / $\mathrm{s}\{c_0\}$	-1.68 $\mathrm{Bq\,m^{-3}}$ / 0.71 $\mathrm{Bq\,m^{-3}}$
Fitted parameter c_∞ / $\mathrm{s}\{c_\infty\}$	25.05 $\mathrm{Bq\,m^{-3}}$ / 2.34 $\mathrm{Bq\,m^{-3}}$
Fixed parameter λ_{eff}	$1.037 \cdot \lambda$, where $\lambda = 2.09838 \cdot 10^{-6}\ \mathrm{s^{-1}}$
Bound-to-free exhalation correction ($J_{\mathrm{M,f}}/J_{\mathrm{M}}$)	1.013

		Conditioning		Build-up		Unit	Notes
Start		201197-14:41		211197-17:53			
Stop		211197-17:53		261197-11:49			
Period			1.13		4.75	d	
Q_{wet}			500		0	$\mathrm{mL_n\,min^{-1}}$	Mass-flow controlled
Q=Q_{wet}+Q_{dry}			1045.0		0	$\mathrm{mL\,min^{-1}}$	Measured at 1014 hPa and 23 °C
c	$\mathrm{s}\{c\}$	-0.3	0.9	7.4	0.6	$\mathrm{Bq\,m^{-3}}$	
T	$\mathrm{s}\{T\}$	20.3	0.1	21.7	0.0	°C	
RH	s{RH}	37.2	0.8	42.0	0.0	%	
P_{atm}	$\mathrm{s}\{P_{\mathrm{atm}}\}$	1012.0	0.5	1018.6	0.2	hPa	
N		27		114		-	Number of measurements

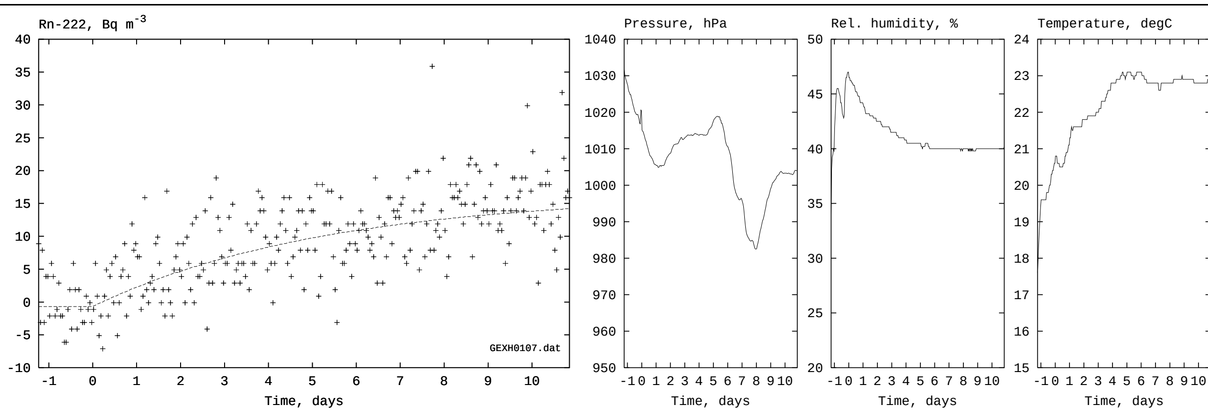


Notes: -

^{222}Rn exhalation rate measurement

Method (primary)	Closed-chamber method w. continuous radon monitor
Method (secondary)	Open-chamber method w. continuous radon monitor
Measurement procedure	Conditioning and build-up (June 1998 procedure)
Measurement no. / Series / Sample ID	#0107 / H+H Industri A/S / M9
Sample descriptor	Bricks
Chamber / Carrier gas	RnChamber2 / nitrogen
Radon instrument / Cycle time	AlphaGuard PQ2000 EF-231 / 1 h
Analysis program / Datafile / Graphfile	`ExhBas02.pas` / `ExhData4.dat` / `GEXH0107.dat`
Date of reporting	August 20, 1998
Experimenter	Claus E. Andersen, Risø National Lab., Denmark
Free mass-specific exhalation rate $J_{\mathrm{M,f}}$	0.10 atoms $\mathrm{s}^{-1}\,\mathrm{kg}^{-1}$
Combined uncertainty ($k=1$) $\mathrm{U_c}\{J_{\mathrm{M,f}}\}$	0.01 atoms $\mathrm{s}^{-1}\,\mathrm{kg}^{-1}$
Type A + B uncert. except Rn monitor bias $\mathrm{u}\{J_{\mathrm{M,f}}\}$	0.01 atoms $\mathrm{s}^{-1}\,\mathrm{kg}^{-1}$
Free area-specific exhalation rate, $J_{\mathrm{A,f}}$	0.020 $\mathrm{Bq\,h^{-1}\,m^{-2}}$
Combined uncertainty ($k=1$) $\mathrm{U_c}\{J_{\mathrm{A,f}}\}$	0.003 $\mathrm{Bq\,h^{-1}\,m^{-2}}$
Type A + B uncert. except Rn monitor bias $\mathrm{u}\{J_{\mathrm{A,f}}\}$	0.003 $\mathrm{Bq\,h^{-1}\,m^{-2}}$
Open-chamber result $J_{\mathrm{M,f,OC}}$ / $\mathrm{u}\{J_{\mathrm{M,f,OC}}\}$	-0.38 atoms $\mathrm{s}^{-1}\,\mathrm{kg}^{-1}$/ 0.71 atoms $\mathrm{s}^{-1}\,\mathrm{kg}^{-1}$
Sample dimension	5 bricks, 3:10x20x5.4cm, 2:10x14.8x5.4cm
Mass (before) / Mass (after) / Mass (lost)	8.5551 kg / 8.5554 kg / -0.3 g
Volume (V) / Area (A) / Density (ρ_{m})	4.84 L / 0.330 m^2/ 1768 $\mathrm{kg\,m^{-3}}$
Empty chamber vol. / dead space ($-$sample) / air	55.76 L / 1.55 L / 49.37 L
Model equation	$c(t) = c_0 + c_\infty(1 - \exp(-\lambda_{\mathrm{eff}} t))$
Chi-2 reduced (χ^2_ν) / N	0.661 / $30 + 260 = 290$
Fitted parameter c_0 / $\mathrm{s}\{c_0\}$	-0.67 $\mathrm{Bq\,m^{-3}}$ / 0.63 $\mathrm{Bq\,m^{-3}}$
Fitted parameter c_∞ / $\mathrm{s}\{c_\infty\}$	17.10 $\mathrm{Bq\,m^{-3}}$ / 1.15 $\mathrm{Bq\,m^{-3}}$
Fixed parameter λ_{eff}	$1.037 \cdot \lambda$, where $\lambda = 2.09838 \cdot 10^{-6}\ \mathrm{s}^{-1}$
Bound-to-free exhalation correction ($J_{\mathrm{M,f}}/J_{\mathrm{M}}$)	1.013

		Conditioning		Build-up		Unit	Notes
Start		171297-11:15		181297-17:32			
Stop		181297-17:32		291297-14:00			
Period			1.26		10.85	d	
Q_{wet}			500		0	$\mathrm{mL_n\,min^{-1}}$	Mass-flow controlled
Q=Q_{wet}+Q_{dry}			1034.0		0	$\mathrm{mL\,min^{-1}}$	Measured at 1017 hPa and 21 °C
c	$\mathrm{s}\{c\}$	0.0	0.7	10.0	0.4	$\mathrm{Bq\,m^{-3}}$	
T	$\mathrm{s}\{T\}$	19.7	0.1	22.4	0.0	°C	
RH	s{RH}	43.5	0.6	41.1	0.1	%	
P_{atm}	$\mathrm{s}\{P_{\mathrm{atm}}\}$	1023.2	0.8	1005.2	0.6	hPa	
N		30		260		-	Number of measurements

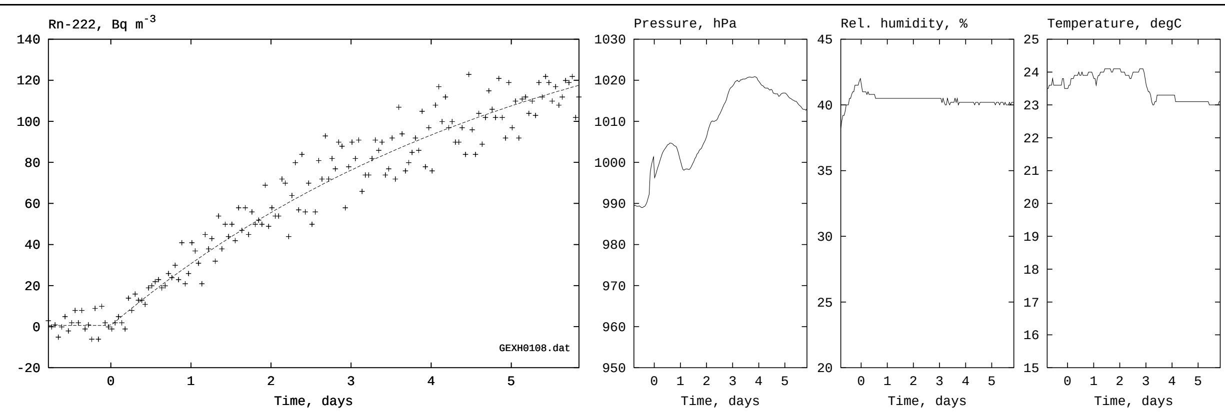


Notes: -

^{222}Rn exhalation rate measurement

Method (primary)	Closed-chamber method w. continuous radon monitor
Method (secondary)	Open-chamber method w. continuous radon monitor
Measurement procedure	Conditioning and build-up (June 1998 procedure)
Measurement no. / Series / Sample ID	#0108 / H+H Industri A/S / M6
Sample descriptor	AAC, density 735
Chamber / Carrier gas	RnChamber2 / nitrogen
Radon instrument / Cycle time	AlphaGuard PQ2000 EF-231 / 1 h
Analysis program / Datafile / Graphfile	`ExhBas02.pas` / `ExhData4.dat` / `GEXH0108.dat`
Date of reporting	August 20, 1998
Experimenter	Claus E. Andersen, Risø National Lab., Denmark
Free mass-specific exhalation rate $J_{\mathrm{M,f}}$	2.50 atoms $\mathrm{s^{-1}\,kg^{-1}}$
Combined uncertainty ($k=1$) $\mathrm{U_c}\{J_{\mathrm{M,f}}\}$	0.16 atoms $\mathrm{s^{-1}\,kg^{-1}}$
Type A + B uncert. except Rn monitor bias $\mathrm{u}\{J_{\mathrm{M,f}}\}$	0.09 atoms $\mathrm{s^{-1}\,kg^{-1}}$
Free area-specific exhalation rate, $J_{\mathrm{A,f}}$	0.286 Bq $\mathrm{h^{-1}\,m^{-2}}$
Combined uncertainty ($k=1$) $\mathrm{U_c}\{J_{\mathrm{A,f}}\}$	0.018 Bq $\mathrm{h^{-1}\,m^{-2}}$
Type A + B uncert. except Rn monitor bias $\mathrm{u}\{J_{\mathrm{A,f}}\}$	0.010 Bq $\mathrm{h^{-1}\,m^{-2}}$
Open-chamber result $J_{\mathrm{M,f,OC}}$ / $\mathrm{u}\{J_{\mathrm{M,f,OC}}\}$	3.45 atoms $\mathrm{s^{-1}\,kg^{-1}}$/ 1.78 atoms $\mathrm{s^{-1}\,kg^{-1}}$
Sample dimension	Slab 29.9 cm x 30.1 cm x 5.1 cm
Mass (before) / Mass (after) / Mass (lost)	3.6543 kg / 3.6570 kg / −2.7 g
Volume (V) / Area (A) / Density (ρ_{m})	4.59 L / 0.241 $\mathrm{m^2}$/ 796 kg $\mathrm{m^{-3}}$
Empty chamber vol. / dead space (−sample) / air	55.76 L / 1.55 L / 49.62 L
Model equation	$c(t) = c_0 + c_\infty(1 - \exp(-\lambda_{\mathrm{eff}} t))$
Chi-2 reduced (χ^2_ν) / N	0.487 / $19 + 141 = 160$
Fitted parameter c_0 / $\mathrm{s}\{c_0\}$	0.58 Bq $\mathrm{m^{-3}}$ / 0.93 Bq $\mathrm{m^{-3}}$
Fitted parameter c_∞ / $\mathrm{s}\{c_\infty\}$	175.47 Bq $\mathrm{m^{-3}}$ / 3.78 Bq $\mathrm{m^{-3}}$
Fixed parameter λ_{eff}	$1.037 \cdot \lambda$, where $\lambda = 2.09838 \cdot 10^{-6}\ \mathrm{s^{-1}}$
Bound-to-free exhalation correction ($J_{\mathrm{M,f}}/J_{\mathrm{M}}$)	1.013

		Conditioning		Build-up		Unit	Notes
Start		050198-08:01		060198-12:43			
Stop		060198-12:43		120198-09:05			
Period			1.20		5.85	d	
Q_{wet}			500		0	$\mathrm{mL_n\,min^{-1}}$	Mass-flow controlled
Q=Q_{wet}+Q_{dry}			1051.0		0	$\mathrm{mL\,min^{-1}}$	Measured at 997 hPa and 25 °C
c	s{c}	1.5	1.1	70.5	2.9	Bq $\mathrm{m^{-3}}$	
T	s{T}	23.6	0.0	23.6	0.0	°C	
RH	s{RH}	40.4	0.3	40.4	0.0	%	
P_{atm}	s{P_{atm}}	991.8	1.0	1011.2	0.7	hPa	
N		19		141		-	Number of measurements

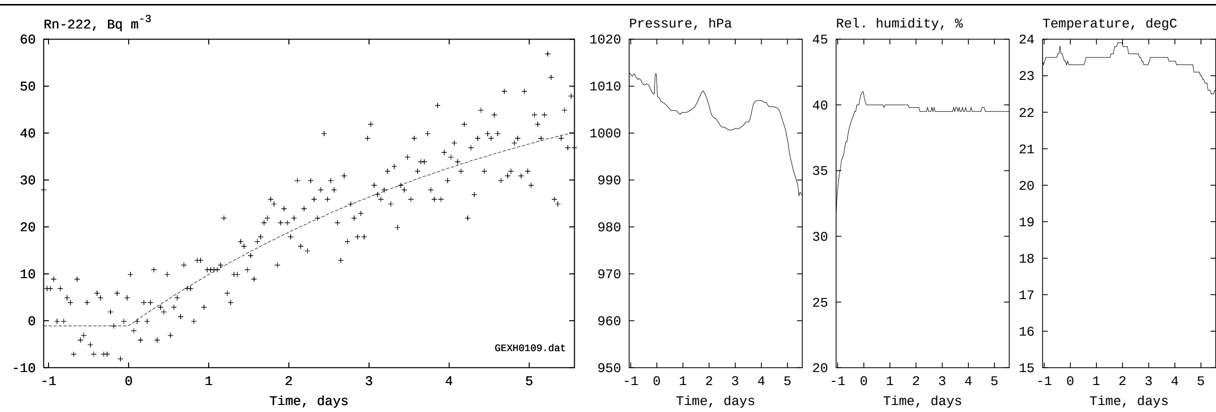


Notes: -

^{222}Rn exhalation rate measurement

Method (primary)	Closed-chamber method w. continuous radon monitor
Method (secondary)	Open-chamber method w. continuous radon monitor
Measurement procedure	Conditioning and build-up (June 1998 procedure)
Measurement no. / Series / Sample ID	#0109 / H+H Industri A/S / M5
Sample descriptor	AAC, density 650
Chamber / Carrier gas	RnChamber2 / nitrogen
Radon instrument / Cycle time	AlphaGuard PQ2000 EF-231 / 1 h
Analysis program / Datafile / Graphfile	`ExhBas02.pas` / `ExhData4.dat` / `GEXH0109.dat`
Date of reporting	August 20, 1998
Experimenter	Claus E. Andersen, Risø National Lab., Denmark
Free mass-specific exhalation rate $J_{\mathrm{M,f}}$	1.06 atoms $\mathrm{s^{-1}\,kg^{-1}}$
Combined uncertainty ($k=1$) $\mathrm{U_c}\{J_{\mathrm{M,f}}\}$	0.09 atoms $\mathrm{s^{-1}\,kg^{-1}}$
Type A + B uncert. except Rn monitor bias $\mathrm{u}\{J_{\mathrm{M,f}}\}$	0.06 atoms $\mathrm{s^{-1}\,kg^{-1}}$
Free area-specific exhalation rate, $J_{\mathrm{A,f}}$	0.103 $\mathrm{Bq\,h^{-1}\,m^{-2}}$
Combined uncertainty ($k=1$) $\mathrm{U_c}\{J_{\mathrm{A,f}}\}$	0.008 $\mathrm{Bq\,h^{-1}\,m^{-2}}$
Type A + B uncert. except Rn monitor bias $\mathrm{u}\{J_{\mathrm{A,f}}\}$	0.006 $\mathrm{Bq\,h^{-1}\,m^{-2}}$
Open-chamber result $J_{\mathrm{M,f,OC}}$ / $\mathrm{u}\{J_{\mathrm{M,f,OC}}\}$	0.18 atoms $\mathrm{s^{-1}\,kg^{-1}}$/ 2.04 atoms $\mathrm{s^{-1}\,kg^{-1}}$
Sample dimension	Slab 30 cm x 30.1 cm x 5.1 cm
Mass (before) / Mass (after) / Mass (lost)	3.1182 kg / 3.1215 kg / −3.3 g
Volume (V) / Area (A) / Density (ρ_{m})	4.61 L / 0.242 $\mathrm{m^2}$/ 677 $\mathrm{kg\,m^{-3}}$
Empty chamber vol. / dead space (−sample) / air	55.76 L / 1.55 L / 49.60 L
Model equation	$c(t) = c_0 + c_\infty(1 - \exp(-\lambda_{\mathrm{eff}} t))$
Chi-2 reduced (χ^2_ν) / N	$0.734 \;/\; 25 + 134 = 159$
Fitted parameter c_0 / $\mathrm{s}\{c_0\}$	−1.05 $\mathrm{Bq\,m^{-3}}$ / 0.77 $\mathrm{Bq\,m^{-3}}$
Fitted parameter c_∞ / $\mathrm{s}\{c_\infty\}$	63.58 $\mathrm{Bq\,m^{-3}}$ / 2.60 $\mathrm{Bq\,m^{-3}}$
Fixed parameter λ_{eff}	$1.037 \cdot \lambda$, where $\lambda = 2.09838 \cdot 10^{-6}\ \mathrm{s^{-1}}$
Bound-to-free exhalation correction ($J_{\mathrm{M,f}}/J_{\mathrm{M}}$)	1.013

		Conditioning		Build-up		Unit	Notes
Start		120198-18:00		130198-18:27			
Stop		130198-18:27		190198-08:59			
Period			1.02		5.61	d	
Q_{wet}			500		0	$\mathrm{mL_n\,min^{-1}}$	Mass-flow controlled
Q=Q_{wet}+Q_{dry}			1037.0		0	$\mathrm{mL\,min^{-1}}$	Measured at 1008 hPa and 25 °C
c	s{c}	1.0	1.2	24.1	1.2	$\mathrm{Bq\,m^{-3}}$	
T	s{T}	23.5	0.0	23.4	0.0	°C	
RH	s{RH}	38.0	0.5	39.7	0.0	%	
P_{atm}	s{P_{atm}}	1011.0	0.3	1003.0	0.4	hPa	
N		25		134		-	Number of measurements

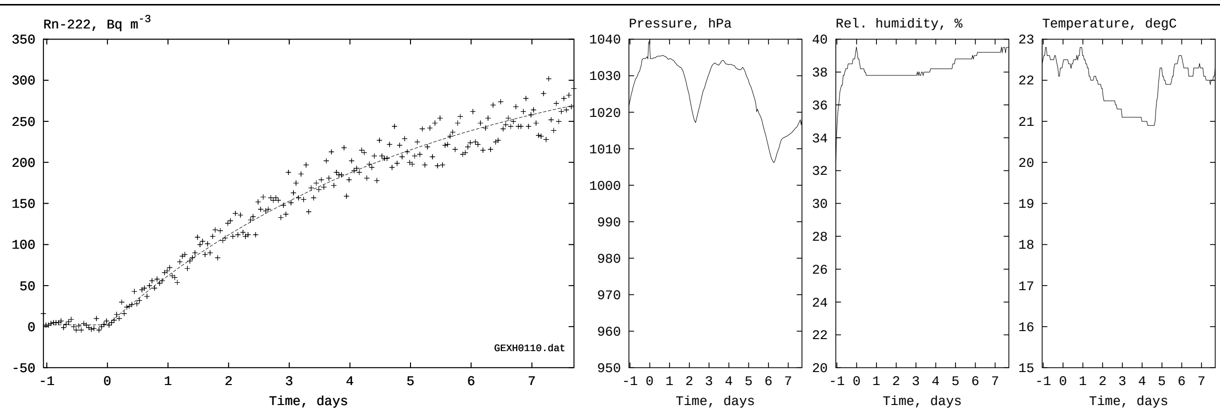


Notes: -

^{222}Rn exhalation rate measurement

Method (primary)	Closed-chamber method w. continuous radon monitor
Method (secondary)	Open-chamber method w. continuous radon monitor
Measurement procedure	Conditioning and build-up (June 1998 procedure)
Measurement no. / Series / Sample ID	#0110 / H+H Industri A/S / M3
Sample descriptor	LAC type 2, density 1500
Chamber / Carrier gas	RnChamber2 / nitrogen
Radon instrument / Cycle time	AlphaGuard PQ2000 EF-231 / 1 h
Analysis program / Datafile / Graphfile	`ExhBas02.pas` / `ExhData4.dat` / `GEXH0110.dat`
Date of reporting	August 20, 1998
Experimenter	Claus E. Andersen, Risø National Lab., Denmark
Free mass-specific exhalation rate $J_{\mathrm{M,f}}$	2.60 atoms s^{-1} kg^{-1}
Combined uncertainty ($k=1$) $\mathrm{U_c}\{J_{\mathrm{M,f}}\}$	0.15 atoms s^{-1} kg^{-1}
Type A + B uncert. except Rn monitor bias $\mathrm{u}\{J_{\mathrm{M,f}}\}$	0.07 atoms s^{-1} kg^{-1}
Free area-specific exhalation rate, $J_{\mathrm{A,f}}$	0.580 Bq h^{-1} m^{-2}
Combined uncertainty ($k=1$) $\mathrm{U_c}\{J_{\mathrm{A,f}}\}$	0.034 Bq h^{-1} m^{-2}
Type A + B uncert. except Rn monitor bias $\mathrm{u}\{J_{\mathrm{A,f}}\}$	0.016 Bq h^{-1} m^{-2}
Open-chamber result $J_{\mathrm{M,f,OC}}$ / $\mathrm{u}\{J_{\mathrm{M,f,OC}}\}$	2.49 atoms s^{-1} kg^{-1}/ 0.90 atoms s^{-1} kg^{-1}
Sample dimension	Slab 29.8 cm x 29.9 cm x 5.0 cm
Mass (before) / Mass (after) / Mass (lost)	7.0345 kg / 7.0365 kg / −2.0 g
Volume (V) / Area (A) / Density (ρ_{m})	4.46 L / 0.238 m^2/ 1579 kg m^{-3}
Empty chamber vol. / dead space (−sample) / air	55.76 L / 1.55 L / 49.75 L
Model equation	$c(t) = c_0 + c_\infty(1 - \exp(-\lambda_{\mathrm{eff}} t))$
Chi-2 reduced (χ^2_ν) / N	0.496 / $26 + 185 = 211$
Fitted parameter c_0 / $\mathrm{s}\{c_0\}$	2.16 Bq m^{-3} / 0.92 Bq m^{-3}
Fitted parameter c_∞ / $\mathrm{s}\{c_\infty\}$	349.69 Bq m^{-3} / 3.79 Bq m^{-3}
Fixed parameter λ_{eff}	$1.037 \cdot \lambda$, where $\lambda = 2.09838 \cdot 10^{-6}$ s^{-1}
Bound-to-free exhalation correction ($J_{\mathrm{M,f}}/J_{\mathrm{M}}$)	1.013

		Conditioning		Build-up		Unit	Notes
Start			200198-16:40		210198-19:20		
Stop			210198-19:20		290198-12:47		
Period			1.11		7.73	d	
Q_{wet}			500		0	$\mathrm{mL_n}$ min^{-1}	Mass-flow controlled
Q=Q_{wet}+Q_{dry}			1031.0		0	mL min^{-1}	Measured at 1035 hPa and 24 °C
c	s{c}	2.6	1.0	168.5	5.6	Bq m^{-3}	
T	s{T}	22.5	0.0	21.9	0.0	°C	
RH	s{RH}	37.6	0.3	38.4	0.0	%	
P_{atm}	s{P_{atm}}	1030.9	1.0	1025.4	0.7	hPa	
N		26		185		-	Number of measurements

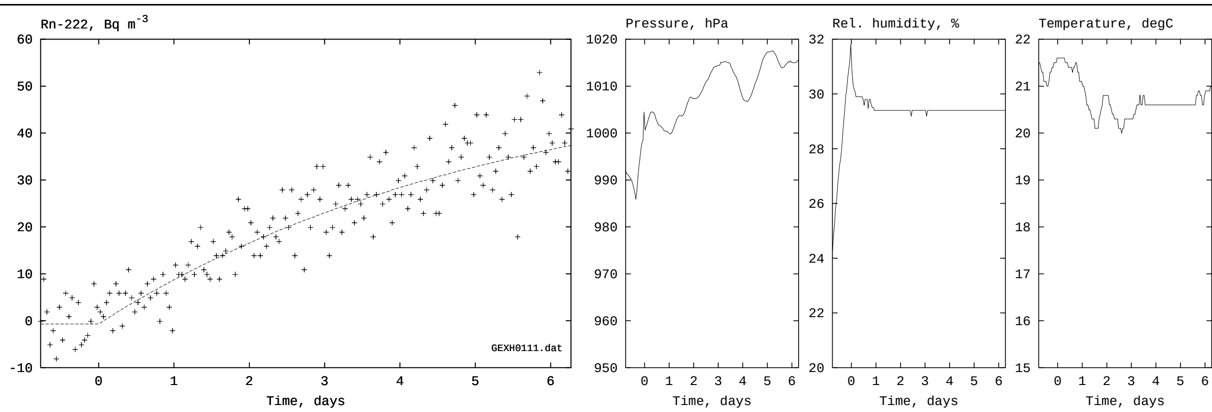


Notes: -

^{222}Rn exhalation rate measurement

Method (primary)	Closed-chamber method w. continuous radon monitor
Method (secondary)	Open-chamber method w. continuous radon monitor
Measurement procedure	Conditioning and build-up (June 1998 procedure)
Measurement no. / Series / Sample ID	#0111 / H+H Industri A/S / M4
Sample descriptor	AAC, density 450
Chamber / Carrier gas	RnChamber2 / nitrogen
Radon instrument / Cycle time	AlphaGuard PQ2000 EF-231 / 1 h
Analysis program / Datafile / Graphfile	`ExhBas02.pas` / `ExhData4.dat` / `GEXH0111.dat`
Date of reporting	August 20, 1998
Experimenter	Claus E. Andersen, Risø National Lab., Denmark
Free mass-specific exhalation rate $J_{\mathrm{M,f}}$	1.26 atoms $\mathrm{s}^{-1}\,\mathrm{kg}^{-1}$
Combined uncertainty ($k=1$) $\mathrm{U_c}\{J_{\mathrm{M,f}}\}$	0.11 atoms $\mathrm{s}^{-1}\,\mathrm{kg}^{-1}$
Type A + B uncert. except Rn monitor bias $\mathrm{u}\{J_{\mathrm{M,f}}\}$	0.08 atoms $\mathrm{s}^{-1}\,\mathrm{kg}^{-1}$
Free area-specific exhalation rate, $J_{\mathrm{A,f}}$	0.087 $\mathrm{Bq\,h^{-1}\,m^{-2}}$
Combined uncertainty ($k=1$) $\mathrm{U_c}\{J_{\mathrm{A,f}}\}$	0.007 $\mathrm{Bq\,h^{-1}\,m^{-2}}$
Type A + B uncert. except Rn monitor bias $\mathrm{u}\{J_{\mathrm{A,f}}\}$	0.006 $\mathrm{Bq\,h^{-1}\,m^{-2}}$
Open-chamber result $J_{\mathrm{M,f,OC}}$ / $\mathrm{u}\{J_{\mathrm{M,f,OC}}\}$	0.30 atoms $\mathrm{s}^{-1}\,\mathrm{kg}^{-1}$/ 2.66 atoms $\mathrm{s}^{-1}\,\mathrm{kg}^{-1}$
Sample dimension	Slab 30.1 cm x 30.0 cm x 5.5 cm
Mass (before) / Mass (after) / Mass (lost)	2.2533 kg / 2.2600 kg / −6.7 g
Volume (V) / Area (A) / Density (ρ_{m})	4.97 L / 0.247 m^2/ 454 $\mathrm{kg\,m^{-3}}$
Empty chamber vol. / dead space (−sample) / air	55.76 L / 1.55 L / 49.24 L
Model equation	$c(t) = c_0 + c_\infty(1 - \exp(-\lambda_{\mathrm{eff}}t))$
Chi-2 reduced (χ^2_ν) / N	0.505 / $19 + 151 = 170$
Fitted parameter c_0 / $\mathrm{s}\{c_0\}$	−0.67 $\mathrm{Bq\,m^{-3}}$ / 0.83 $\mathrm{Bq\,m^{-3}}$
Fitted parameter c_∞ / $\mathrm{s}\{c_\infty\}$	54.92 $\mathrm{Bq\,m^{-3}}$ / 2.38 $\mathrm{Bq\,m^{-3}}$
Fixed parameter λ_{eff}	$1.037 \cdot \lambda$, where $\lambda = 2.09838 \cdot 10^{-6}\ \mathrm{s}^{-1}$
Bound-to-free exhalation correction ($J_{\mathrm{M,f}}/J_{\mathrm{M}}$)	1.013

		Conditioning		Build-up		Unit	Notes
Start		020298-14:25		030298-15:30			
Stop		030298-15:30		090298-22:28			
Period			1.05		6.29	d	
Q_{wet}			500		0	$\mathrm{mL_n\,min^{-1}}$	Mass-flow controlled
Q=Q_{wet}+Q_{dry}			982.4		0	$\mathrm{mL\,min^{-1}}$	Measured at 1001 hPa and 23 °C
c	$\mathrm{s}\{c\}$	0.1	1.2	22.9	1.0	$\mathrm{Bq\,m^{-3}}$	
T	$\mathrm{s}\{T\}$	21.3	0.0	20.7	0.0	°C	
RH	s{RH}	28.1	0.5	29.5	0.0	%	
P_{atm}	$\mathrm{s}\{P_{\mathrm{atm}}\}$	992.1	1.1	1009.7	0.4	hPa	
N		19		151		-	Number of measurements

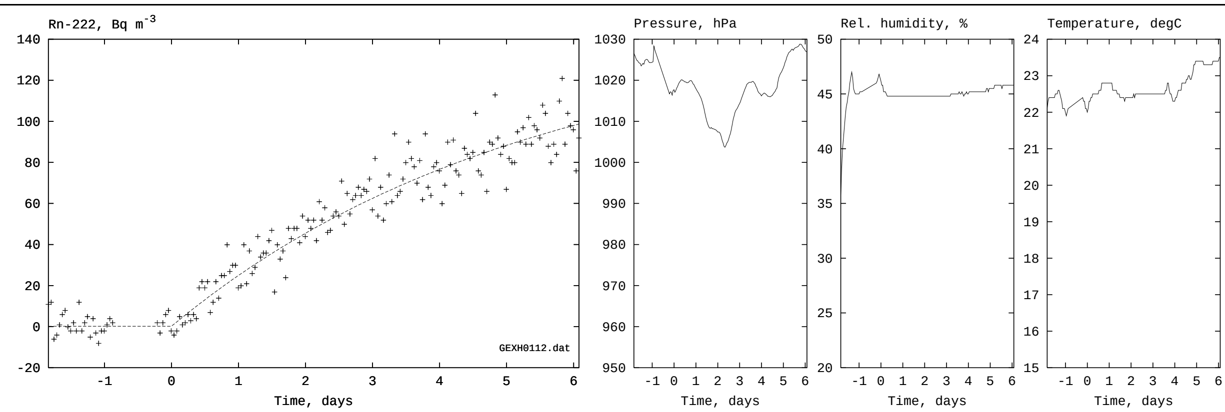


Notes: -

^{222}Rn exhalation rate measurement

Method (primary)	Closed-chamber method w. continuous radon monitor
Method (secondary)	Open-chamber method w. continuous radon monitor
Measurement procedure	Conditioning and build-up (June 1998 procedure)
Measurement no. / Series / Sample ID	#0112 / H+H Industri A/S / M1
Sample descriptor	LAC, density 600
Chamber / Carrier gas	RnChamber2 / nitrogen
Radon instrument / Cycle time	AlphaGuard PQ2000 EF-231 / 1 h
Analysis program / Datafile / Graphfile	`ExhBas02.pas` / `ExhData4.dat` / `GEXH0112.dat`
Date of reporting	August 20, 1998
Experimenter	Claus E. Andersen, Risø National Lab., Denmark
Free mass-specific exhalation rate $J_{\mathrm{M,f}}$	2.62 atoms $\mathrm{s}^{-1}\,\mathrm{kg}^{-1}$
Combined uncertainty ($k=1$) $\mathrm{U_c}\{J_{\mathrm{M,f}}\}$	0.17 atoms $\mathrm{s}^{-1}\,\mathrm{kg}^{-1}$
Type A + B uncert. except Rn monitor bias $\mathrm{u}\{J_{\mathrm{M,f}}\}$	0.10 atoms $\mathrm{s}^{-1}\,\mathrm{kg}^{-1}$
Free area-specific exhalation rate, $J_{\mathrm{A,f}}$	0.239 Bq $\mathrm{h}^{-1}\,\mathrm{m}^{-2}$
Combined uncertainty ($k=1$) $\mathrm{U_c}\{J_{\mathrm{A,f}}\}$	0.015 Bq $\mathrm{h}^{-1}\,\mathrm{m}^{-2}$
Type A + B uncert. except Rn monitor bias $\mathrm{u}\{J_{\mathrm{A,f}}\}$	0.009 Bq $\mathrm{h}^{-1}\,\mathrm{m}^{-2}$
Open-chamber result $J_{\mathrm{M,f,OC}}$ / $\mathrm{u}\{J_{\mathrm{M,f,OC}}\}$	3.14 atoms $\mathrm{s}^{-1}\,\mathrm{kg}^{-1}$/ 2.00 atoms $\mathrm{s}^{-1}\,\mathrm{kg}^{-1}$
Sample dimension	Slab 30.0 cm x 30.0 cm x 4.9 cm
Mass (before) / Mass (after) / Mass (lost)	2.8890 kg / 2.8925 kg / −3.5 g
Volume (V) / Area (A) / Density (ρ_{m})	4.41 L / 0.239 m^2/ 655 $\mathrm{kg\,m}^{-3}$
Empty chamber vol. / dead space (−sample) / air	55.76 L / 1.55 L / 49.80 L
Model equation	$c(t) = c_0 + c_\infty(1 - \exp(-\lambda_{\mathrm{eff}} t))$
Chi-2 reduced (χ^2_ν) / N	0.609 / $30 + 146 = 176$
Fitted parameter c_0 / $\mathrm{s}\{c_0\}$	0.16 $\mathrm{Bq\,m}^{-3}$ / 0.80 $\mathrm{Bq\,m}^{-3}$
Fitted parameter c_∞ / $\mathrm{s}\{c_\infty\}$	144.64 $\mathrm{Bq\,m}^{-3}$ / 3.25 $\mathrm{Bq\,m}^{-3}$
Fixed parameter λ_{eff}	$1.037 \cdot \lambda$, where $\lambda = 2.09838 \cdot 10^{-6}\ \mathrm{s}^{-1}$
Bound-to-free exhalation correction ($J_{\mathrm{M,f}}/J_{\mathrm{M}}$)	1.013

		Conditioning		Build-up		Unit	Notes
Start			170298-20:10		190298-17:06		
Stop			190298-17:06		250298-19:30		
Period			1.87		6.10	d	
Q_{wet}			500		0	$\mathrm{mL_n\,min}^{-1}$	Mass-flow controlled
$Q = Q_{\mathrm{wet}} + Q_{\mathrm{dry}}$			965.1		0	$\mathrm{mL\,min}^{-1}$	Measured at 1017 hPa and 23 °C
c	$\mathrm{s}\{c\}$	1.4	1.0	60.0	2.4	$\mathrm{Bq\,m}^{-3}$	
T	$\mathrm{s}\{T\}$	22.3	0.0	22.7	0.0	°C	
RH	s{RH}	44.5	0.5	45.1	0.0	%	
P_{atm}	$\mathrm{s}\{P_{\mathrm{atm}}\}$	1023.4	0.6	1017.1	0.5	hPa	
N		30		146		-	Number of measurements

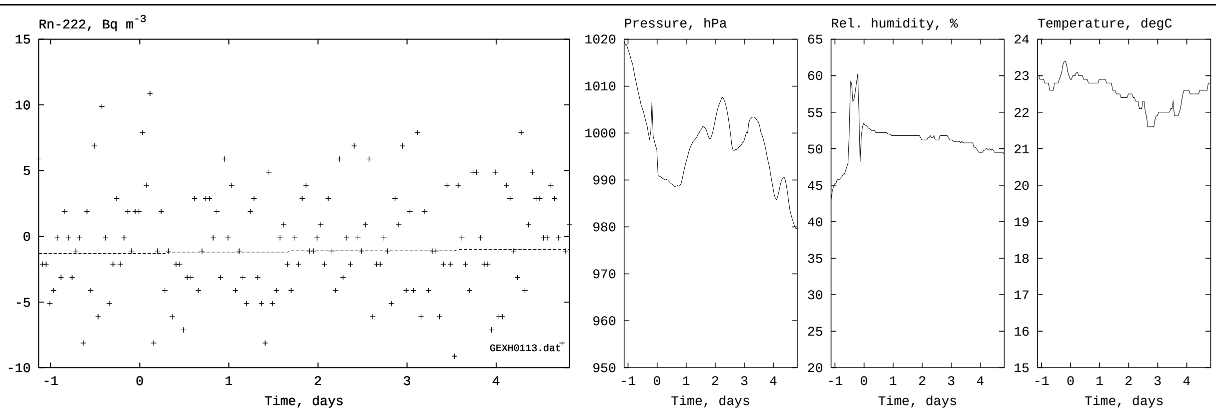


Notes: Initial build-up period (180298-21:15 to 190298-09:49) abandoned because of ill-closed chamber .

^{222}Rn exhalation rate measurement

Method (primary)	Closed-chamber method w. continuous radon monitor
Method (secondary)	Open-chamber method w. continuous radon monitor
Measurement procedure	Conditioning and build-up (June 1998 procedure)
Measurement no. / Series / Sample ID	#0113 / H+H Industri A/S / M10
Sample descriptor	Lightw. expand. clay agg.
Chamber / Carrier gas	RnChamber2 / nitrogen
Radon instrument / Cycle time	AlphaGuard PQ2000 EF-231 / 1 h
Analysis program / Datafile / Graphfile	`ExhBas02.pas` / `ExhData4.dat` / `GEXH0113.dat`
Date of reporting	August 20, 1998
Experimenter	Claus E. Andersen, Risø National Lab., Denmark
Free mass-specific exhalation rate $J_{\mathrm{M,f}}$	0.02 atoms $\mathrm{s}^{-1}\,\mathrm{kg}^{-1}$
Combined uncertainty ($k=1$) $\mathrm{U_c}\{J_{\mathrm{M,f}}\}$	0.10 atoms $\mathrm{s}^{-1}\,\mathrm{kg}^{-1}$
Type A + B uncert. except Rn monitor bias $\mathrm{u}\{J_{\mathrm{M,f}}\}$	0.10 atoms $\mathrm{s}^{-1}\,\mathrm{kg}^{-1}$
Free area-specific exhalation rate, $J_{\mathrm{A,f}}$	0.001 Bq $\mathrm{h}^{-1}\,\mathrm{m}^{-2}$
Combined uncertainty ($k=1$) $\mathrm{U_c}\{J_{\mathrm{A,f}}\}$	0.004 Bq $\mathrm{h}^{-1}\,\mathrm{m}^{-2}$
Type A + B uncert. except Rn monitor bias $\mathrm{u}\{J_{\mathrm{A,f}}\}$	0.004 Bq $\mathrm{h}^{-1}\,\mathrm{m}^{-2}$
Open-chamber result $J_{\mathrm{M,f,OC}}$ / $\mathrm{u}\{J_{\mathrm{M,f,OC}}\}$	−2.69 atoms $\mathrm{s}^{-1}\,\mathrm{kg}^{-1}$/ 3.60 atoms $\mathrm{s}^{-1}\,\mathrm{kg}^{-1}$
Sample dimension	Single grains (no packing)
Mass (before) / Mass (after) / Mass (lost)	1.5086 kg / 1.5085 kg / 0.1 g
Volume (V) / Area (A) / Density (ρ_{m})	5.19 L / 0.277 m^2/ 291 kg m^{-3}
Empty chamber vol. / dead space (−sample) / air	55.76 L / 1.55 L / 49.02 L
Model equation	$c(t) = c_0 + c_\infty(1 - \exp(-\lambda_{\mathrm{eff}}t))$
Chi-2 reduced (χ^2_ν) / N	0.638 / $28 + 116 = 144$
Fitted parameter c_0 / $\mathrm{s}\{c_0\}$	−1.28 Bq m^{-3} / 0.70 Bq m^{-3}
Fitted parameter c_∞ / $\mathrm{s}\{c_\infty\}$	0.46 Bq m^{-3} / 2.03 Bq m^{-3}
Fixed parameter λ_{eff}	$1.037 \cdot \lambda$, where $\lambda = 2.09838 \cdot 10^{-6}\ \mathrm{s}^{-1}$
Bound-to-free exhalation correction ($J_{\mathrm{M,f}}/J_{\mathrm{M}}$)	1.013

		Conditioning		Build-up		Unit	Notes
Start			260298-10:20		270298-20:14		
Stop			270298-20:14		040398-16:52		
Period			1.41		4.86	d	
Q_{wet}			500		0	$\mathrm{mL_n\ min^{-1}}$	Mass-flow controlled
Q=Q_{wet}+Q_{dry}			926.4		0	$\mathrm{mL\ min^{-1}}$	Measured at 992 hPa and 25 °C
c	$\mathrm{s}\{c\}$	−0.6	0.8	−0.5	0.4	Bq m^{-3}	
T	$\mathrm{s}\{T\}$	22.9	0.0	22.4	0.0	°C	
RH	s{RH}	50.4	1.1	51.2	0.1	%	
P_{atm}	$\mathrm{s}\{P_{\mathrm{atm}}\}$	1007.9	1.4	995.3	0.6	hPa	
N		28		116		-	Number of measurements

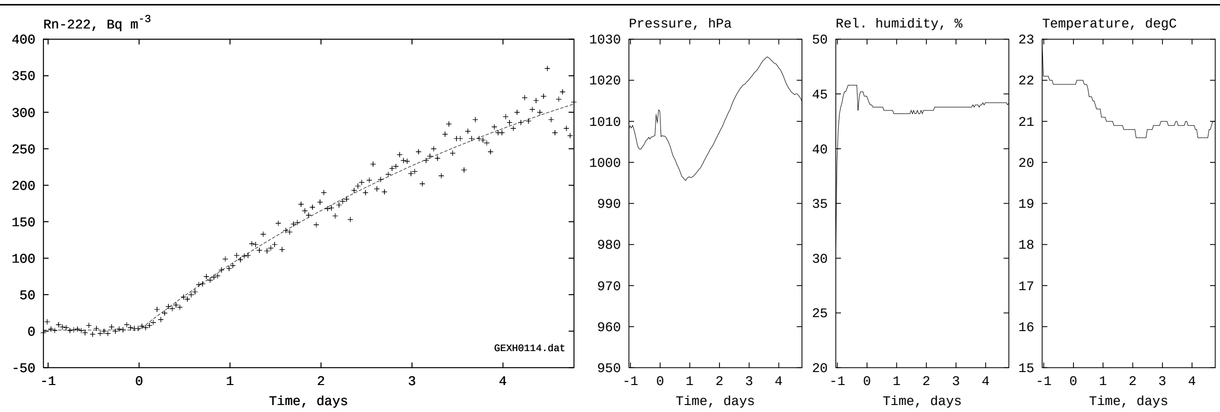


Notes: -

^{222}Rn exhalation rate measurement

Method (primary)	Closed-chamber method w. continuous radon monitor
Method (secondary)	Open-chamber method w. continuous radon monitor
Measurement procedure	Conditioning and build-up (June 1998 procedure)
Measurement no. / Series / Sample ID	#0114 / H+H Industri A/S / M7
Sample descriptor	Ordinary concrete, density 2300
Chamber / Carrier gas	RnChamber2 / nitrogen
Radon instrument / Cycle time	AlphaGuard PQ2000 EF-231 / 1 h
Analysis program / Datafile / Graphfile	`ExhBas02.pas` / `ExhData4.dat` / `GEXH0114.dat`
Date of reporting	August 20, 1998
Experimenter	Claus E. Andersen, Risø National Lab., Denmark
Free mass-specific exhalation rate $J_{\mathrm{M,f}}$	2.70 atoms $\mathrm{s^{-1}\,kg^{-1}}$
Combined uncertainty ($k=1$) $\mathrm{U_c}\{J_{\mathrm{M,f}}\}$	0.16 atoms $\mathrm{s^{-1}\,kg^{-1}}$
Type A + B uncert. except Rn monitor bias $\mathrm{u}\{J_{\mathrm{M,f}}\}$	0.07 atoms $\mathrm{s^{-1}\,kg^{-1}}$
Free area-specific exhalation rate, $J_{\mathrm{A,f}}$	0.861 Bq $\mathrm{h^{-1}\,m^{-2}}$
Combined uncertainty ($k=1$) $\mathrm{U_c}\{J_{\mathrm{A,f}}\}$	0.050 Bq $\mathrm{h^{-1}\,m^{-2}}$
Type A + B uncert. except Rn monitor bias $\mathrm{u}\{J_{\mathrm{A,f}}\}$	0.024 Bq $\mathrm{h^{-1}\,m^{-2}}$
Open-chamber result $J_{\mathrm{M,f,OC}}$ / $\mathrm{u}\{J_{\mathrm{M,f,OC}}\}$	2.20 atoms $\mathrm{s^{-1}\,kg^{-1}}$/ 0.67 atoms $\mathrm{s^{-1}\,kg^{-1}}$
Sample dimension	Slab 29.9 cm x 30.0 cm x 5.0 cm
Mass (before) / Mass (after) / Mass (lost)	10.0843 kg / 10.0844 kg / −0.1 g
Volume (V) / Area (A) / Density (ρ_{m})	4.49 L / 0.239 $\mathrm{m^2}$/ 2248 $\mathrm{kg\,m^{-3}}$
Empty chamber vol. / dead space (−sample) / air	55.76 L / 1.55 L / 49.72 L
Model equation	$c(t) = c_0 + c_\infty(1 - \exp(-\lambda_{\mathrm{eff}} t))$
Chi-2 reduced (χ^2_ν) / N	0.456 / $26 + 115 = 141$
Fitted parameter c_0 / $\mathrm{s}\{c_0\}$	1.60 Bq $\mathrm{m^{-3}}$ / 0.97 Bq $\mathrm{m^{-3}}$
Fitted parameter c_∞ / $\mathrm{s}\{c_\infty\}$	522.21 Bq $\mathrm{m^{-3}}$ / 6.63 Bq $\mathrm{m^{-3}}$
Fixed parameter λ_{eff}	$1.037 \cdot \lambda$, where $\lambda = 2.09838 \cdot 10^{-6}\ \mathrm{s^{-1}}$
Bound-to-free exhalation correction ($J_{\mathrm{M,f}}/J_{\mathrm{M}}$)	1.013

		Conditioning		Build-up		Unit	Notes
Start		050398-19:42		060398-21:16			
Stop		060398-21:16		110398-16:08			
Period			1.07		4.79	d	
Q_{wet}			500		0	$\mathrm{mL_n\,min^{-1}}$	Mass-flow controlled
$Q{=}Q_{\mathrm{wet}}{+}Q_{\mathrm{dry}}$			1074.0		0	$\mathrm{mL\,min^{-1}}$	Measured at 1007 hPa and 23 °C
c	$\mathrm{s}\{c\}$	2.8	0.8	181.1	8.6	$\mathrm{Bq\,m^{-3}}$	
T	$\mathrm{s}\{T\}$	22.0	0.0	21.0	0.0	°C	
RH	s{RH}	44.2	0.6	43.7	0.0	%	
P_{atm}	$\mathrm{s}\{P_{\mathrm{atm}}\}$	1006.8	0.5	1011.4	0.9	hPa	
N		26		115		-	Number of measurements

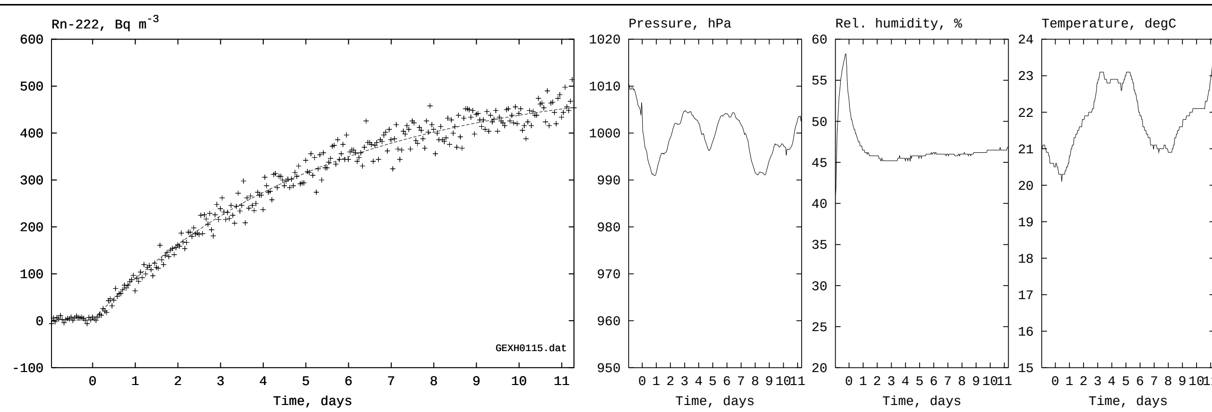


Notes: -

^{222}Rn exhalation rate measurement

Method (primary)	Closed-chamber method w. continuous radon monitor
Method (secondary)	Open-chamber method w. continuous radon monitor
Measurement procedure	Conditioning and build-up (June 1998 procedure)
Measurement no. / Series / Sample ID	#0115 / H+H Industri A/S / M7
Sample descriptor	Ordinary concrete, density 2300
Chamber / Carrier gas	RnChamber2 / nitrogen
Radon instrument / Cycle time	AlphaGuard PQ2000 EF-231 / 1 h
Analysis program / Datafile / Graphfile	`ExhBas02.pas` / `ExhData4.dat` / `GEXH0115.dat`
Date of reporting	August 20, 1998
Experimenter	Claus E. Andersen, Risø National Lab., Denmark
Free mass-specific exhalation rate $J_{\mathrm{M,f}}$	2.67 atoms $\mathrm{s}^{-1}\,\mathrm{kg}^{-1}$
Combined uncertainty ($k=1$) $\mathrm{U_c}\{J_{\mathrm{M,f}}\}$	0.15 atoms $\mathrm{s}^{-1}\,\mathrm{kg}^{-1}$
Type A + B uncert. except Rn monitor bias $\mathrm{u}\{J_{\mathrm{M,f}}\}$	0.07 atoms $\mathrm{s}^{-1}\,\mathrm{kg}^{-1}$
Free area-specific exhalation rate, $J_{\mathrm{A,f}}$	0.849 $\mathrm{Bq\,h^{-1}\,m^{-2}}$
Combined uncertainty ($k=1$) $\mathrm{U_c}\{J_{\mathrm{A,f}}\}$	0.048 $\mathrm{Bq\,h^{-1}\,m^{-2}}$
Type A + B uncert. except Rn monitor bias $\mathrm{u}\{J_{\mathrm{A,f}}\}$	0.021 $\mathrm{Bq\,h^{-1}\,m^{-2}}$
Open-chamber result $J_{\mathrm{M,f,OC}}$ / $\mathrm{u}\{J_{\mathrm{M,f,OC}}\}$	2.74 atoms $\mathrm{s}^{-1}\,\mathrm{kg}^{-1}$/ 0.61 atoms $\mathrm{s}^{-1}\,\mathrm{kg}^{-1}$
Sample dimension	Slab 29.9 cm x 30.0 cm x 5.0 cm
Mass (before) / Mass (after) / Mass (lost)	10.0754 kg / 10.0790 kg / −3.6 g
Volume (V) / Area (A) / Density (ρ_{m})	4.49 L / 0.239 $\mathrm{m^2}$/ 2247 $\mathrm{kg\,m^{-3}}$
Empty chamber vol. / dead space (−sample) / air	55.76 L / 1.55 L / 49.72 L
Model equation	$c(t) = c_0 + c_\infty(1 - \exp(-\lambda_{\mathrm{eff}} t))$
Chi-2 reduced (χ^2_ν) / N	0.551 / $24 + 270 = 294$
Fitted parameter c_0 / $\mathrm{s}\{c_0\}$	1.41 $\mathrm{Bq\,m^{-3}}$ / 0.99 $\mathrm{Bq\,m^{-3}}$
Fitted parameter c_∞ / $\mathrm{s}\{c_\infty\}$	515.09 $\mathrm{Bq\,m^{-3}}$ / 3.44 $\mathrm{Bq\,m^{-3}}$
Fixed parameter λ_{eff}	$1.037 \cdot \lambda$, where $\lambda = 2.09838 \cdot 10^{-6}\ \mathrm{s}^{-1}$
Bound-to-free exhalation correction ($J_{\mathrm{M,f}}/J_{\mathrm{M}}$)	1.013

		Conditioning		Build-up		Unit	Notes
Start			020498-10:00		030498-14:07		
Stop			030498-14:07		140498-21:00		
Period			1.17		11.29	d	
Q_{wet}			500		0	$\mathrm{mL_n\,min^{-1}}$	Mass-flow controlled
$Q{=}Q_{\mathrm{wet}}{+}Q_{\mathrm{dry}}$			915.8		0	$\mathrm{mL\,min^{-1}}$	Measured at 1001 hPa and 22 °C
c	$\mathrm{s}\{c\}$	3.8	1.0	304.1	7.9	$\mathrm{Bq\,m^{-3}}$	
T	$\mathrm{s}\{T\}$	20.8	0.0	21.9	0.1	°C	
RH	s{RH}	53.3	1.0	46.2	0.1	%	
P_{atm}	$\mathrm{s}\{P_{\mathrm{atm}}\}$	1007.8	0.4	998.6	0.3	hPa	
N		24		270		-	Number of measurements

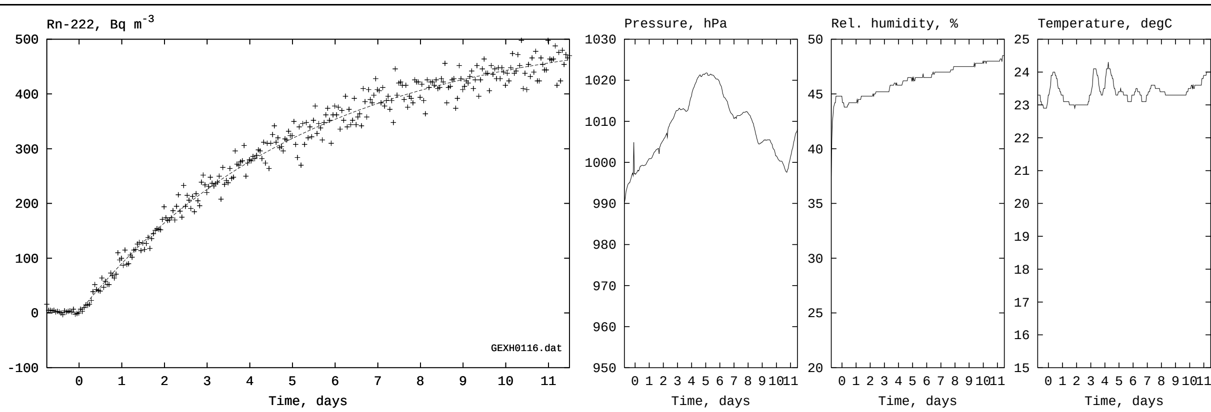


Notes: -

^{222}Rn exhalation rate measurement

Method (primary)	Closed-chamber method w. continuous radon monitor
Method (secondary)	Open-chamber method w. continuous radon monitor
Measurement procedure	Conditioning and build-up (June 1998 procedure)
Measurement no. / Series / Sample ID	#0116 / H+H Industri A/S / M7
Sample descriptor	Ordinary concrete, density 2300
Chamber / Carrier gas	RnChamber2 / nitrogen
Radon instrument / Cycle time	AlphaGuard PQ2000 EF-231 / 1 h
Analysis program / Datafile / Graphfile	`ExhBas02.pas` / `ExhData4.dat` / `GEXH0116.dat`
Date of reporting	August 20, 1998
Experimenter	Claus E. Andersen, Risø National Lab., Denmark
Free mass-specific exhalation rate $J_{\mathrm{M,f}}$	2.70 atoms $\mathrm{s^{-1}\,kg^{-1}}$
Combined uncertainty ($k=1$) $\mathrm{U_c}\{J_{\mathrm{M,f}}\}$	0.15 atoms $\mathrm{s^{-1}\,kg^{-1}}$
Type A + B uncert. except Rn monitor bias $\mathrm{u}\{J_{\mathrm{M,f}}\}$	0.07 atoms $\mathrm{s^{-1}\,kg^{-1}}$
Free area-specific exhalation rate, $J_{\mathrm{A,f}}$	0.859 $\mathrm{Bq\,h^{-1}\,m^{-2}}$
Combined uncertainty ($k=1$) $\mathrm{U_c}\{J_{\mathrm{A,f}}\}$	0.049 $\mathrm{Bq\,h^{-1}\,m^{-2}}$
Type A + B uncert. except Rn monitor bias $\mathrm{u}\{J_{\mathrm{A,f}}\}$	0.022 $\mathrm{Bq\,h^{-1}\,m^{-2}}$
Open-chamber result $J_{\mathrm{M,f,OC}}$ / $\mathrm{u}\{J_{\mathrm{M,f,OC}}\}$	1.55 atoms $\mathrm{s^{-1}\,kg^{-1}}$/ 0.68 atoms $\mathrm{s^{-1}\,kg^{-1}}$
Sample dimension	Slab 29.9 cm x 30.0 cm x 5.0 cm
Mass (before) / Mass (after) / Mass (lost)	10.0777 kg / 10.0779 kg / −0.2 g
Volume (V) / Area (A) / Density (ρ_{m})	4.49 L / 0.239 $\mathrm{m^2}$/ 2247 $\mathrm{kg\,m^{-3}}$
Empty chamber vol. / dead space (−sample) / air	55.76 L / 1.55 L / 49.72 L
Model equation	$c(t) = c_0 + c_\infty(1-\exp(-\lambda_{\mathrm{eff}}t))$
Chi-2 reduced (χ^2_ν) / N	0.420 / $19+276=295$
Fitted parameter c_0 / $\mathrm{s}\{c_0\}$	1.23 $\mathrm{Bq\,m^{-3}}$ / 1.04 $\mathrm{Bq\,m^{-3}}$
Fitted parameter c_∞ / $\mathrm{s}\{c_\infty\}$	521.16 $\mathrm{Bq\,m^{-3}}$ / 3.45 $\mathrm{Bq\,m^{-3}}$
Fixed parameter λ_{eff}	$1.037\cdot\lambda$, where $\lambda = 2.09838\cdot10^{-6}\ \mathrm{s^{-1}}$
Bound-to-free exhalation correction ($J_{\mathrm{M,f}}/J_{\mathrm{M}}$)	1.013

		Conditioning		Build-up		Unit	Notes
Start			160498-18:12		170498-13:12		
Stop			170498-13:12		290498-09:50		
Period			0.79		11.86	d	
Q_{wet}			500		0	$\mathrm{mL_n\,min^{-1}}$	Mass-flow controlled
$Q=Q_{\mathrm{wet}}+Q_{\mathrm{dry}}$			1125.0		0	$\mathrm{mL\,min^{-1}}$	Measured at 997 hPa and 25 °C
c	$\mathrm{s}\{c\}$	2.9	1.0	310.5	7.8	$\mathrm{Bq\,m^{-3}}$	
T	$\mathrm{s}\{T\}$	23.1	0.0	23.4	0.0	°C	
RH	s{RH}	43.8	0.4	46.4	0.1	%	
P_{atm}	$\mathrm{s}\{P_{\mathrm{atm}}\}$	995.4	0.7	1009.3	0.4	hPa	
N		19		276		-	Number of measurements

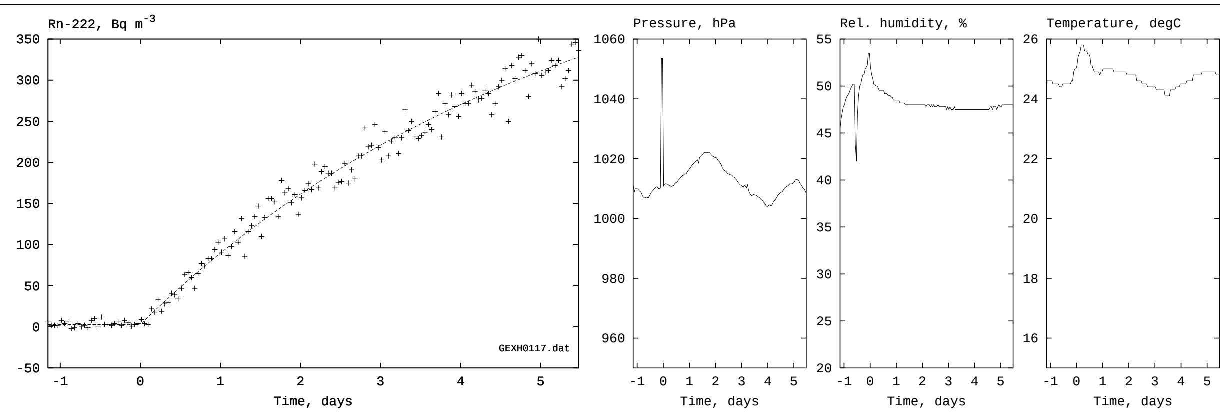


Notes: -

^{222}Rn exhalation rate measurement

Method (primary)	Closed-chamber method w. continuous radon monitor
Method (secondary)	Open-chamber method w. continuous radon monitor
Measurement procedure	Conditioning and build-up (June 1998 procedure)
Measurement no. / Series / Sample ID	#0117 / H+H Industri A/S / M7
Sample descriptor	Ordinary concrete, density 2300
Chamber / Carrier gas	RnChamber2 / nitrogen
Radon instrument / Cycle time	AlphaGuard PQ2000 EF-231 / 1 h
Analysis program / Datafile / Graphfile	`ExhBas02.pas` / `ExhData4.dat` / `GEXH0117.dat`
Date of reporting	August 20, 1998
Experimenter	Claus E. Andersen, Risø National Lab., Denmark
Free mass-specific exhalation rate $J_{\mathrm{M,f}}$	2.62 atoms $\mathrm{s^{-1}\,kg^{-1}}$
Combined uncertainty ($k=1$) $\mathrm{U_c}\{J_{\mathrm{M,f}}\}$	0.15 atoms $\mathrm{s^{-1}\,kg^{-1}}$
Type A + B uncert. except Rn monitor bias $\mathrm{u}\{J_{\mathrm{M,f}}\}$	0.07 atoms $\mathrm{s^{-1}\,kg^{-1}}$
Free area-specific exhalation rate, $J_{\mathrm{A,f}}$	0.834 $\mathrm{Bq\,h^{-1}\,m^{-2}}$
Combined uncertainty ($k=1$) $\mathrm{U_c}\{J_{\mathrm{A,f}}\}$	0.048 $\mathrm{Bq\,h^{-1}\,m^{-2}}$
Type A + B uncert. except Rn monitor bias $\mathrm{u}\{J_{\mathrm{A,f}}\}$	0.022 $\mathrm{Bq\,h^{-1}\,m^{-2}}$
Open-chamber result $J_{\mathrm{M,f,OC}}$ / $\mathrm{u}\{J_{\mathrm{M,f,OC}}\}$	3.23 atoms $\mathrm{s^{-1}\,kg^{-1}}$/ 0.73 atoms $\mathrm{s^{-1}\,kg^{-1}}$
Sample dimension	Slab 29.9 cm x 30.0 cm x 5.0 cm
Mass (before) / Mass (after) / Mass (lost)	10.0711 kg / 10.0722 kg / −1.1 g
Volume (V) / Area (A) / Density (ρ_{m})	4.49 L / 0.239 $\mathrm{m^2}$/ 2246 $\mathrm{kg\,m^{-3}}$
Empty chamber vol. / dead space (−sample) / air	55.76 L / 1.55 L / 49.72 L
Model equation	$c(t) = c_0 + c_\infty(1 - \exp(-\lambda_{\mathrm{eff}} t))$
Chi-2 reduced (χ^2_ν) / N	0.456 / 28 + 131 = 159
Fitted parameter c_0 / $\mathrm{s}\{c_0\}$	2.82 $\mathrm{Bq\,m^{-3}}$ / 0.91 $\mathrm{Bq\,m^{-3}}$
Fitted parameter c_∞ / $\mathrm{s}\{c_\infty\}$	505.90 $\mathrm{Bq\,m^{-3}}$ / 5.77 $\mathrm{Bq\,m^{-3}}$
Fixed parameter λ_{eff}	$1.037 \cdot \lambda$, where $\lambda = 2.09838 \cdot 10^{-6}\ \mathrm{s^{-1}}$
Bound-to-free exhalation correction ($J_{\mathrm{M,f}}/J_{\mathrm{M}}$)	1.013

		Conditioning		Build-up		Unit	Notes
Start		020698-16:15		030698-21:40			
Stop		030698-21:40		090698-09:00			
Period			1.23		5.47	d	
Q_{wet}			500		0	$\mathrm{mL_n\,min^{-1}}$	Mass-flow controlled
Q=Q_{wet}+Q_{dry}			1125.0		0	$\mathrm{mL\,min^{-1}}$	Measured at 997 hPa and 25 °C
c	$\mathrm{s}\{c\}$	3.6	0.6	192.9	8.3	$\mathrm{Bq\,m^{-3}}$	
T	$\mathrm{s}\{T\}$	24.6	0.0	24.7	0.0	°C	
RH	s{RH}	49.2	0.5	48.1	0.1	%	
P_{atm}	$\mathrm{s}\{P_{\mathrm{atm}}\}$	1012.1	2.3	1012.8	0.4	hPa	
N		28		131		-	Number of measurements

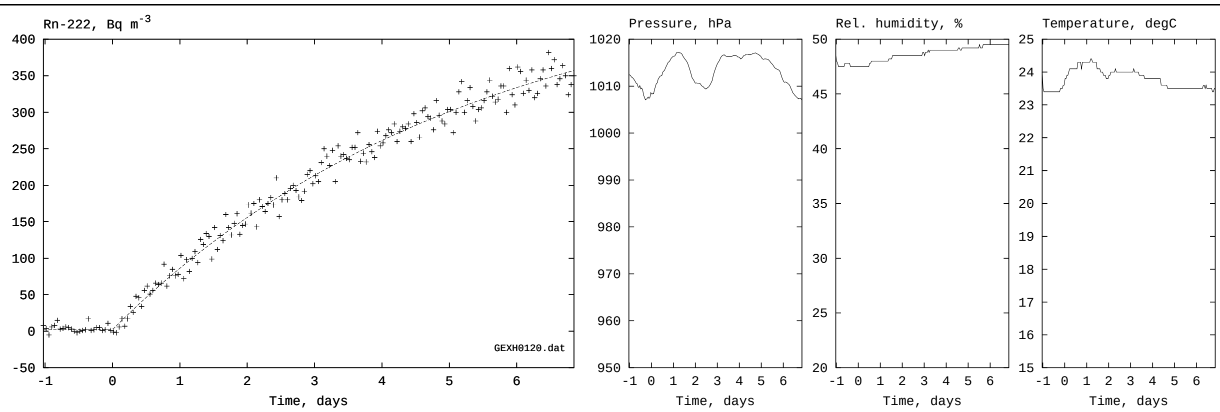


Notes: -

^{222}Rn exhalation rate measurement

Method (primary)	Closed-chamber method w. continuous radon monitor
Method (secondary)	Open-chamber method w. continuous radon monitor
Measurement procedure	Conditioning and build-up (June 1998 procedure)
Measurement no. / Series / Sample ID	#0120 / H+H Industri A/S / M7
Sample descriptor	Ordinary concrete, density 2300
Chamber / Carrier gas	RnChamber2 / nitrogen
Radon instrument / Cycle time	AlphaGuard PQ2000 EF-231 / 1 h
Analysis program / Datafile / Graphfile	`ExhBas02.pas` / `ExhData4.dat` / `GEXH0120.dat`
Date of reporting	August 20, 1998
Experimenter	Claus E. Andersen, Risø National Lab., Denmark
Free mass-specific exhalation rate $J_{\mathrm{M,f}}$	2.54 atoms $\mathrm{s}^{-1}\,\mathrm{kg}^{-1}$
Combined uncertainty ($k=1$) $\mathrm{U_c}\{J_{\mathrm{M,f}}\}$	0.15 atoms $\mathrm{s}^{-1}\,\mathrm{kg}^{-1}$
Type A + B uncert. except Rn monitor bias $\mathrm{u}\{J_{\mathrm{M,f}}\}$	0.07 atoms $\mathrm{s}^{-1}\,\mathrm{kg}^{-1}$
Free area-specific exhalation rate, $J_{\mathrm{A,f}}$	0.808 Bq $\mathrm{h}^{-1}\,\mathrm{m}^{-2}$
Combined uncertainty ($k=1$) $\mathrm{U_c}\{J_{\mathrm{A,f}}\}$	0.046 Bq $\mathrm{h}^{-1}\,\mathrm{m}^{-2}$
Type A + B uncert. except Rn monitor bias $\mathrm{u}\{J_{\mathrm{A,f}}\}$	0.021 Bq $\mathrm{h}^{-1}\,\mathrm{m}^{-2}$
Open-chamber result $J_{\mathrm{M,f,OC}}$ / $\mathrm{u}\{J_{\mathrm{M,f,OC}}\}$	3.67 atoms $\mathrm{s}^{-1}\,\mathrm{kg}^{-1}$/ 0.76 atoms $\mathrm{s}^{-1}\,\mathrm{kg}^{-1}$
Sample dimension	Slab 29.9 cm x 30.0 cm x 5.0 cm
Mass (before) / Mass (after) / Mass (lost)	10.0716 kg / 10.0714 kg / 0.2 g
Volume (V) / Area (A) / Density (ρ_{m})	4.49 L / 0.239 m^2/ 2246 kg m^{-3}
Empty chamber vol. / dead space (−sample) / air	55.76 L / 1.55 L / 49.72 L
Model equation	$c(t) = c_0 + c_\infty(1 - \exp(-\lambda_{\mathrm{eff}}t))$
Chi-2 reduced (χ^2_ν) / N	0.453 / $25 + 165 = 190$
Fitted parameter c_0 / $\mathrm{s}\{c_0\}$	2.17 Bq m^{-3} / 0.97 Bq m^{-3}
Fitted parameter c_∞ / $\mathrm{s}\{c_\infty\}$	490.16 Bq m^{-3} / 4.82 Bq m^{-3}
Fixed parameter λ_{eff}	$1.037 \cdot \lambda$, where $\lambda = 2.09838 \cdot 10^{-6}\ \mathrm{s}^{-1}$
Bound-to-free exhalation correction ($J_{\mathrm{M,f}}/J_{\mathrm{M}}$)	1.013

		Conditioning		Build-up		Unit	Notes
Start		200798-17:30		210798-18:37			
Stop		210798-18:37		280798-15:33			
Period			1.05		6.87	d	
Q_{wet}			500		0	$\mathrm{mL_n\,min^{-1}}$	Mass-flow controlled
$Q{=}Q_{\mathrm{wet}}{+}Q_{\mathrm{dry}}$			1118.0		0	$\mathrm{mL\,min^{-1}}$	Measured at 1009 hPa and 26 °C
c	$\mathrm{s}\{c\}$	4.0	1.0	217.7	8.0	$\mathrm{Bq\,m^{-3}}$	
T	$\mathrm{s}\{T\}$	23.4	0.0	23.8	0.0	°C	
RH	s{RH}	47.6	0.0	48.8	0.0	%	
P_{atm}	$\mathrm{s}\{P_{\mathrm{atm}}\}$	1009.7	0.4	1013.6	0.2	hPa	
N		25		165		-	Number of measurements

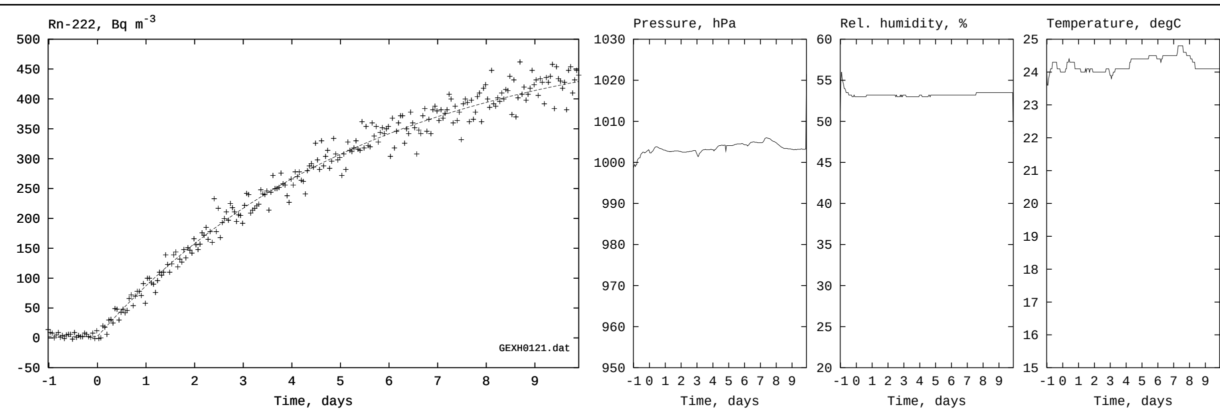


Notes: -

^{222}Rn exhalation rate measurement

Method (primary)	Closed-chamber method w. continuous radon monitor
Method (secondary)	Open-chamber method w. continuous radon monitor
Measurement procedure	Conditioning and build-up (June 1998 procedure)
Measurement no. / Series / Sample ID	#0121 / H+H Industri A/S / M7
Sample descriptor	Ordinary concrete, density 2300
Chamber / Carrier gas	RnChamber2 / nitrogen
Radon instrument / Cycle time	AlphaGuard PQ2000 EF-231 / 1 h
Analysis program / Datafile / Graphfile	`ExhBas02.pas` / `ExhData4.dat` / `GEXH0121.dat`
Date of reporting	August 20, 1998
Experimenter	Claus E. Andersen, Risø National Lab., Denmark
Free mass-specific exhalation rate $J_{\mathrm{M,f}}$	2.56 atoms $\mathrm{s^{-1}\,kg^{-1}}$
Combined uncertainty ($k=1$) $\mathrm{U_c}\{J_{\mathrm{M,f}}\}$	0.15 atoms $\mathrm{s^{-1}\,kg^{-1}}$
Type A + B uncert. except Rn monitor bias $\mathrm{u}\{J_{\mathrm{M,f}}\}$	0.06 atoms $\mathrm{s^{-1}\,kg^{-1}}$
Free area-specific exhalation rate, $J_{\mathrm{A,f}}$	0.814 $\mathrm{Bq\,h^{-1}\,m^{-2}}$
Combined uncertainty ($k=1$) $\mathrm{U_c}\{J_{\mathrm{A,f}}\}$	0.046 $\mathrm{Bq\,h^{-1}\,m^{-2}}$
Type A + B uncert. except Rn monitor bias $\mathrm{u}\{J_{\mathrm{A,f}}\}$	0.021 $\mathrm{Bq\,h^{-1}\,m^{-2}}$
Open-chamber result $J_{\mathrm{M,f,OC}}$ / $\mathrm{u}\{J_{\mathrm{M,f,OC}}\}$	3.14 atoms $\mathrm{s^{-1}\,kg^{-1}}$/ 0.66 atoms $\mathrm{s^{-1}\,kg^{-1}}$
Sample dimension	Slab 29.9 cm x 30.0 cm x 5.0 cm
Mass (before) / Mass (after) / Mass (lost)	10.0729 kg / 10.0727 kg / 0.2 g
Volume (V) / Area (A) / Density (ρ_{m})	4.49 L / 0.239 $\mathrm{m^2}$/ 2246 $\mathrm{kg\,m^{-3}}$
Empty chamber vol. / dead space (−sample) / air	55.76 L / 1.55 L / 49.72 L
Model equation	$c(t) = c_0 + c_\infty(1 - \exp(-\lambda_{\mathrm{eff}} t))$
Chi-2 reduced (χ^2_ν) / N	0.476 / $25 + 237 = 262$
Fitted parameter c_0 / $\mathrm{s}\{c_0\}$	2.30 $\mathrm{Bq\,m^{-3}}$ / 0.98 $\mathrm{Bq\,m^{-3}}$
Fitted parameter c_∞ / $\mathrm{s}\{c_\infty\}$	511.88 $\mathrm{Bq\,m^{-3}}$ / 3.78 $\mathrm{Bq\,m^{-3}}$
Fixed parameter λ_{eff}	$1.000 \cdot \lambda$, where $\lambda = 2.09838 \cdot 10^{-6}\ \mathrm{s^{-1}}$
Bound-to-free exhalation correction ($J_{\mathrm{M,f}}/J_{\mathrm{M}}$)	1.013

		Conditioning		Build-up		Unit	Notes
Start		300798-10:55		310798-11:20			
Stop		310798-11:20		100898-08:56			
Period			1.02		9.90	d	
Q_{wet}			500		0	$\mathrm{mL_n\,min^{-1}}$	Mass-flow controlled
$Q = Q_{\mathrm{wet}} + Q_{\mathrm{dry}}$			991.9		0	$\mathrm{mL\,min^{-1}}$	Measured at 1005 hPa and 26 °C
c	$\mathrm{s}\{c\}$	4.6	0.9	277.1	8.0	$\mathrm{Bq\,m^{-3}}$	
T	$\mathrm{s}\{T\}$	24.1	0.0	24.2	0.0	°C	
RH	s{RH}	53.6	0.2	53.2	0.0	%	
P_{atm}	$\mathrm{s}\{P_{\mathrm{atm}}\}$	1001.4	0.3	1003.6	0.1	hPa	
N		25		237		-	Number of measurements



Notes: Chamber pressure test is ok.

Bibliographic Data Sheet **Risø-R-1135(EN)**

Title and author(s)

Radon-222 exhalation from Danish building materials: H+H Industri A/S results

Claus E. Andersen

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Pages	Tables	Illustrations	References
56	6	15	29

Abstract (Max. 2000 char.)

This report describes a closed-chamber method for laboratory measurements of the rate at which radon-222 degasses (exhales) from small building material samples. The chamber is 55 L in volume and the main sample geometry is a slab of dimensions 5x30x30 cm^3. Numerical modelling is used to assess (and partly remove) the bias of the method relative to an ideal measurement of the free exhalation rate. Experimental results obtained with the method are found to be in agreement with the results of an open-chamber method (which is subject to different sources of error).

Results of radon-222 exhalation rate measurements for 10 samples of Danish building materials are reported. Samples include ordinary concrete, lightweight aggregate concrete, autoclaved aerated concrete, bricks, and gypsum board. The maximum mass-specific exhalation rate is about 20 $mBq\,h^{-1}\,kg^{-1}$. Under consideration of the specific applications of the investigated building materials, the contribution to the indoor radon-222 concentration in a single-family reference house is calculated. Numerical modelling is used to help extrapolate the laboratory measurements on small samples to full scale walls. Application of typical materials will increase the indoor concentration by less than 10 $Bq\,m^{-3}$.

Descriptors INIS/EDB

BRICKS; CONCRETES; DEGASSING; DIFFUSION; FINITE DIFFERENCE METHOD; GYPSUM CEMENTS; HOUSES; MEASURING METHODS; RADIOECOLOGICAL CONCENTRATIONS; RADIUM 226; RADON 222; RISOE NATIONAL LABORATORY

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